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ENVIRONMENTAL RADIOACTIVITY AT ARGONNE NATIONAL LABORATORY

Report for the Years 1960 and 1961

by

J. Sedlet and F. S. Iwami

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ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60440

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Written by

J. Sedlet and F. S. Iwami

Experimental Work Done by:

C. D. Eldred
N. W. Golchert
C. M. Greene, Jr.
F. S. Iwami
K. K. Kamalick
J. Sedlet

Industrial Hygiene and Safety Division

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ENVIRONMENTAL RADIOACTIVITY AT ARGONNE NATIONAL LABORATORY

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I. SUMMARY

The results of the environmental monitoring program at Argonne National Laboratory for 1960 and 1961 are given in this report. Quarterly and semiannual summaries of some of these results have appeared in Radiological Health Data, Vol. I, No. 9; Vol. II, Nos. 4, 7, and 12; and Vol. III, No. 5. This report presents the complete results of the program. The purposes of the program are to measure the natural radioactive content of the ANL site and its surroundings, and to determine the origin, identity, and magnitude of any radioactivity above the natural levels. Of primary interest is the detection of any radioactive materials released to the environment as a result of the program at Argonne.

The radioactive content of the environment was measured by radiochemical analyses, total activity measurements, and gamma-ray spectrometry of several types of natural materials collected on the ANL site and from locations up to 100 miles from the Laboratory. The frequency of sampling decreased with increasing distance from the site. Since air and water are the most probable media for spreading radioactive contamination, the sampling program has concentrated on these materials. Argonne waste water is discharged into Sawmill Creek, which in turn flows into the Des Plaines River, and special emphasis was placed on these streams.

The average total activities in samples of water, material from the beds of lakes and streams (bottom silt), soil, and plants during 1960 and 1961 are given in Figures 1 through 6. For comparison purposes, the results obtained from 1952 through 1961 are given in Figures 7 through 12.

In 1960 and the first three quarters of 1961, fallout activity in the environment was at the lowest level since January 1953, since there was no large-scale nuclear testing in the atmosphere during 1959 and 1960. The particulate beta activity remaining in air filter samples after the decay of radon and thoron daughters averaged approximately 0.1 pc/m^3, compared to annual averages ranging from 0.5 to 3.0 pc/m^3 between 1953 and 1959. Increases in the total long-lived beta activity and in the concentrations of individual fission products of 20 to 50% were noted from March through July 1960, and during May and June 1961. These increases can be attributed to a "spring maximum" in stratospheric fallout. A corresponding increase in the fall of 1960 was not obvious. Fallout from the French nuclear tests in the Sahara was not sufficiently large to affect the total beta activity in air appreciably, but fallout from these tests was detected in precipitation during February and March 1960.

On September 14, 1961, the beta activity in air increased sharply, and remained at a relatively high level for the remainder of the year, as a result of the series of atmospheric nuclear detonations conducted by the USSR beginning on September 1, 1961. The beta activity from September 14, 1961, to the end of the year averaged 14 pc/m^3, compared with 0.1 pc/m^3 during the first half of September. Fission products from these detonations were also found in other types of materials during the fall and winter of 1961. The fallout was most readily detectable in air, precipitation, and grass. Concentrations of specific fission products in air and precipitation are given in Section III of this report.

Radioactivity originating at Argonne and leaving the site was found only in air during March 1961, and in most Sawmill Creek water samples collected during the reporting period. The airborne activity was due to iodine-131 accidentally released to the atmosphere in the exhaust air from two of the Argonne buildings. Iodine-131 was found in air samples collected on the ANL site during the latter half of February and most of March 1961. In March it was also present at two of the four off-site sampling locations (southwest and southeast of the site) at average monthly concentrations of 0.41 and 0.36 pc/m^3, or 0.14 and 0.12% of the maximum permissible concentration (MPC),* respectively. Iodine-131 was not detected in any of the off-site air samples during February 1961, the limit of detection being 0.035 pc/m^3 (0.012% of the MPC). The average concentrations on the site during February and March were 1.9 and 3.3 pc/m^3, respectively. The maximum concentration in any single air sample on the site was 16.7 pc/m^3 (5.6% of the MPC). The release had essentially ceased by the end of March, and during the remainder of the year only two on-site air samples, one collected in April and one in May, contained detectable amounts of iodine-131 originating at Argonne. The iodine content of these samples was equivalent to 0.006 and 0.011% of the MPC, respectively. Except during March 1961, no significant or consistent differences could be detected between the radioactivity in air samples collected on and off the site.

Iodine-131 from this release was also found in precipitation, grass, and soil on the ANL site and in Sawmill Creek water. The maximum and average iodine-131 concentrations in Sawmill Creek during the period of this release were 55 and 14 pc/l, or 2.8 and 0.7% of the MPC for drinking water. The iodine-131 concentrations in grass on the site during and

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*The maximum permissible concentrations used in this report are those recommended by the National Committee on Radiation Protection (NCRP) in National Bureau of Standards Handbook 69 (June 5, 1959) for uncontrolled areas in the vicinity of a nuclear installation. The values are one-tenth of those specified for an occupational exposure of 168 hr/week. In evaluating the effect of a nuclear installation on its environment, the NCRP states that concentrations of radionuclides may be averaged over periods up to one year. This suggestion is usually followed in this report in making comparisons with the MPC's, although averages over shorter periods and maximum concentrations are used at times when such comparisons are of interest. In the latter instances, the percent of MPC is overestimated. The MPC's given by the NCRP for water are for drinking water only, and although they are applied in this report to Sawmill Creek, it should be pointed out that Sawmill Creek water is not used for human consumption. Generally accepted MPC's for other types of water are not available.

following the release varied from 15 x 10^3 pc/g near the building from which most of the iodine-131 escaped to less than 1 pc/g at the northern and eastern boundaries of the site and to 2 pc/g at the southern and western boundaries. Iodine-131 concentrations in soil were approximately one-hundred times less (per gram) than in grass at the same location. Radioiodine was not detected in soil more than 150 yards from the point of release, while detectable amounts were present in grass 2400 yards from the release. The distribution pattern on and off the site indicated that the bulk of the iodine-131 moved in a southerly direction. Iodine-131 from the release was not found in any grass samples collected off the ANL site, even at locations where it was found in air and as close as 2.5 miles southwest of the site. The air concentrations off the site were apparently too low to result in detectable surface deposition. Iodine-131 from this release was not detected in grass or soil after June 1961. The low concentrations in the off-site air samples and the rapid rate of disappearance from the surface, where surface deposition was detected, indicate that the iodine-131 released by Argonne did not constitute a health hazard.

The concentrations of radioactive materials added to Sawmill Creek by Argonne waste water were determined by comparing the concentrations in Creek water collected above the site and below the waste water outfall. The principal radioactive nuclides added to Sawmill Creek by Argonne waste water were cobalt-58, cobalt-60, uranium and the thorium-protactinium-234 daughters of natural uranium, strontium-90, and cesium-137. The iodine-131 content of Sawmill Creek water from the release early in 1961 was given earlier. The average concentrations of cobalt-58, the predominant beta emitter, were 369 pc/l (0.4% of the MPC) in 1960 and 59 pc/l (0.06% of the MPC) in 1961. Average concentrations of the other beta emitters added by Argonne waste water ranged from 18 pc/l for thorium-234 in 1960 to 1 pc/l for strontium-90 in 1961. In terms of the MPC's, the greatest contribution was made by strontium-90, whose average concentration amounted to 5% of the MPC in 1960 and 1% in 1961. Each of the other beta emitters was present in concentrations equivalent to less than 0.5% of their MPC's.

The alpha activity added to the Creek by Argonne waste water was due almost entirely to normal uranium. The average uranium concentrations below the outfall were 29 pc/l (0.073% of the MPC) in 1960 and 5 pc/l (0.013% of the MPC) in 1961. The natural concentration of uranium in the Creek is about 1.2 pc/l. Small amounts of plutonium and thorium were occasionally added to the Creek in Argonne waste water. The average concentrations of these elements were 0.09 pc/l (0.0017% of the MPC) for plutonium and 0.12 pc/l (0.01% of the MPC) for thorium.

The average total beta activity in below-outfall Creek water during 1960 was 37 pc/l. Approximately 3 pc/l was due to activity naturally present in the Creek, 1 pc/l was due to fallout, and the remainder was added in Argonne waste water. In 1961 the average fallout activity increased to 7 pc/l and the Argonne contribution decreased to 20 pc/l.

The activity added to Sawmill Creek in Argonne waste water was also found in about one-third of the bottom silt samples collected below the outfall. At this location the total alpha activities varied from 19 to 60 pc/g and the beta activities from 34 to 175 pc/g, compared with above-site activities ranging from 16 to 30 pcα/g and from 37 to 73 pcβ/g. The same nuclides added to the Creek in the waste water were found in the bottom silt in abnormally high concentrations.

Two other locations on the ANL site contained radioactive materials originating at Argonne. Water and bottom silt from a lagoon occasionally used for the storage of contaminated waste water contained elevated concentrations of the same nuclides added to Sawmill Creek in Argonne waste water, and surface soil and grass near a shed formerly used for uranium storage contained abnormal amounts of uranium.

The radioactivity in the Des Plaines River was essentially the same both above and below the mouth of Sawmill Creek. This was true for the total alpha and beta activities as well as for uranium and the fission products for which analyses were made. Plutonium, thorium, and the cobalt nuclides were not detected at either location. Apparently as the result of dilution, the activity in Sawmill Creek had no detectable effect on the radioactivity in the Des Plaines River.

The range of alpha activities, 0.1 to 4.7 pc/l, in surface water collected off the ANL site was normal during both years. The beta activities in surface water were relatively low up to September 1961. The average value, about 8 pc/l, is close to the natural level, since the fallout contribution was small. The beta activities in some of the samples collected after September 1961 increased by up to a factor of two as a result of fallout from the USSR test series, and similar increases were noted in bottom silt.

The total activities in soil collected off the ANL site were normal, averaging about 20 pcα/g and 50 pcβ/g. Fission product fallout activity was low compared with the natural activity, even after September 1961, and did not increase the total beta activity appreciably. Several samples were analyzed for uranium, thorium, and plutonium. The uranium content varied from 1.7 to 2.2 pc/g, and the thorium concentrations from 0.016 to 0.021 pc/g. Similar results were obtained in other years. Plutonium concentrations were between 0.03 and 0.05 pc/g. The plutonium presumably originated in fallout.

Plant samples collected in October 1961 contained beta activities about five times greater than samples obtained earlier in 1961 or in 1960. The increase was due to fission products from the USSR test series beginning September 1961. At other times during 1960 and 1961, the beta activities in plants at all sampling locations were relatively low and averaged about 30 pc/g. Except for grass collected near the uranium storage shed, the alpha activities at all locations were in the normal range of 0.2 to 2 pc/g.

II. PROGRAM AND PROCEDURES

The results of the environmental monitoring program at Argonne National Laboratory for 1960 and 1961 are given in this report. Quarterly and semiannual summaries of some of these results have previously appeared in Radiological Health Data, Volume I, No. 9; Volume II, Nos. 4, 7, and 12; and Volume III, No. 5. This report presents the complete results of the program. The purposes of the program are to measure the natural radioactive content of the ANL site and its environment, and to determine the identity and magnitude of any radioactivity above the natural levels. Of primary interest is the detection of any radioactive materials released to the environment by Argonne.

The radioactivity of the environment was determined by radiochemical analyses, total activity measurements, and gamma-ray spectrometry on the types and numbers of samples given in Table I. These samples were collected on the ANL site and from locations up to 100 miles from the site. The sampling locations are shown in Figures 13 and 14. Samples collected near ANL are intended to indicate the direction and extent of contamination in the event significant amounts of radioactive materials are accidentally released to the environment by Argonne. In general, it is expected that natural and fallout activity will be relatively uniform on the site and within 25 miles of the site, while activity released by Argonne should be present in higher concentrations on the site. Thus, higher activities on the site would indicate a release of activity by Argonne, while similar activities on, and within 25 miles of, the site would indicate that no significant release has occurred. Interpretations based on such evidence have usually been valid, although localized differences in fallout and natural activity are sometimes observed, and conclusions regarding the origin of non-natural activity must be reached cautiously.

TABLE I

SAMPLES COLLECTED IN 1960 AND 1961

Type	Number	
	1960	1961
Water	328	351
Precipitation	67	96
Soil	93	81
Bottom Silt	98	90
Plant	60	102
Air Filter	836	906

The samples collected 100 miles from the Laboratory (referred to as "reference sites") were originally intended to serve primarily as continuous checks for contamination during collection, storage, and analysis, since their radioactivity was not expected to change with time. However, because of the widespread occurrence of fallout, the beta activities in these samples have served primarily to indicate the extent and magnitude of fallout activity. Since there is little non-natural alpha activity in the environment, the primary purpose of the reference site samples has been realized

for alpha activity. Since fallout activity was found in all environmental materials in varying amounts, beta activities have been more difficult to interpret in terms of origin than alpha activities. However, it was usually possible to distinguish between fallout, naturally occurring activity, and activity from Argonne operations by making the proper choices of sampling locations and types of analyses.

Measurements of total alpha and beta activity were made by counting thick samples (5 to 75 mg/cm^2) after a minimum of sample preparation.

Water and precipitation samples, acidified with nitric acid after collection to prevent hydrolysis, were evaporated to dryness. Several milligrams of silver iodide were precipitated from the rain and snow samples prior to evaporation to retain trace amounts of radioiodine. The residue was flamed, ground in a mortar, and spread uniformly on a weighed counting planchet with carbon tetrachloride. The residue was counted after it had been dried and the planchet reweighed.

Soil and bottom silt were dried at 110°C, ground in a mortar, and a weighed portion spread on a counting planchet.

Plant samples were washed with water to remove dirt, dried at 110°C, ashed, and a weighed portion mounted for counting.

Animal samples were air dried at room temperature, ashed, and a weighed portion counted.

Air filter samples were sprayed with a solution of polystyrene in ethylene dichloride to fix the dust on the paper prior to mounting the filter paper on a planchet. The polystyrene layer was 0.1 mg/cm^2 thick.

Total alpha counting was done in nylon-window gas-flow proportional counters or zinc sulfide scintillation counters. Beta particles were counted in nylon-window gas-flow proportional counters installed in a lead and anticoincidence shield to reduce the background. The thicknesses of the nylon windows ranged from 0.1 to 0.2 mg/cm^2.

For air filters, the correction factors used to obtain disintegration rates from counting rates were those measured for radon daughters. For the other types of samples, the counting rates were converted to disintegration rates by applying correction factors measured for plutonium-239 (for alpha particles) and thallium-204 (for beta particles). The results obtained in this way represent the true disintegration rates if all the radioactive nuclides in the samples emitted particles of the same energies as the nuclides used in obtaining the correction factors. These types of corrections were used for total alpha and beta counting since the samples were

thick and contained unknown and variable mixtures of radionuclides. True disintegration rates in such samples cannot be obtained by counting total activity alone, and a standardized but arbitrary method for obtaining nominal disintegration rates must be used. However, measurements of total activity were made because large numbers of such analyses can be made rapidly, and the results are very useful in comparing activity levels and in determining which samples should be analyzed further.

Gamma-ray counting was done with a 4 x 4-in. sodium iodide crystal connected to a linear amplifier and a 256-channel analyzer. The crystal was shielded by 6 in. of iron lined with $\frac{1}{8}$ in. of low-activity lead to reduce backscattering. Calibrations of energy and counting efficiency were made with standardized solutions of the appropriate nuclides.

Fission product and radiocobalt analyses were made by separating the desired element with carrier added and counting the activity in anti-coincidence shielded beta counters. Counting rates were converted to disintegration rates by applying correction factors experimentally determined for each nuclide being counted.

Uranium analyses were made with a fluorophotometer, and the results converted to disintegration rates, assuming the natural isotopic composition of uranium.

Plutonium and thorium (including the thorium-234 daughters of uranium) analyses were made by coprecipitation with cerium fluoride, followed by extraction with a solution of thenoyltrifluoroacetone in benzene. In the extraction the two elements were separated from each other and from other activities by adjusting the acidity of the solution and the oxidation state of the plutonium. The separated plutonium and thorium fractions were counted for alpha activity in 2π proportional counters.

Additional details on the sampling program, instrumentation, counting techniques, and radiochemical analyses are given in the previous reports in this series: ANL-5069, -5289, -5446, -5684, -5808, -5934, -6047, and -6282.

III. RADIOACTIVITY IN ENVIRONMENTAL SAMPLES

A. Air Filters

Airborne particulate matter was sampled at six locations on the ANL site and at four locations off the site. Samples were collected by drawing outside air through Hollingsworth and Vose No. 70 filter paper. At one on-site location the filter paper was changed daily to record short-term changes in air activity, whereas at all other locations the papers were changed at weekly intervals. The weekly samples were used primarily to compare off-site and on-site activities. The alpha and beta activities in the weekly on-site samples were determined during the first day after the end of the collection period to obtain the radon and thoron concentrations, and again 3 days (in 1960) or 4 days (in 1961) and 7 days after collection to obtain the long-lived activity. The off-site air samples could not be obtained sufficiently early for the measurements of radon and thoron daughters, and were counted after 3 (or 4) and 7 days. Until September 1961, the daily air samples were counted once, after 3 (in 1960) or 4 (in 1961) days.

After the resumption of nuclear testing in September 1961, the daily samples were counted on the same schedule as the weekly samples. Such a sequence of counting measurements provides sufficient data for a calculation of the age of the fission product activity from the fourth and seventh day counts and for obtaining an approximate value for fallout activity 4 to 6 hr after collection. From the ratio of alpha to beta activities after the radon daughter products have decayed, the contribution of the thoron daughter to the total beta activity can be subtracted, since this ratio is known for samples containing only thoron daughters.

The counting schedule was changed in January 1961 from 3 to 4 days after collection because the amount of decay between 3 and 7 days indicated that a significant fraction of the activity remaining after 3 days was still due to thoron daughter products; consequently, the third day count did not give a correct value for the "long-lived" activity. The relative contribution of the thoron daughter activity to the total activity depends, of course, on the amount of "long-lived" activity, and was much greater for alpha than for beta activity, as can be seen from the results in Tables II and III. The presence of thoron daughters would lower the apparent age of fallout activity when the age was calculated from the third and seventh day beta counts. The ratio of the third to fourth day activities in six weekly samples collected in December 1960 varied from 1.74 to 1.97 for the alpha activity and from 1.06 to 1.16 for the beta activity. In eight daily samples, the corresponding ratio was 1.75 to 4.06 for the alpha activity and 1.09 to 2.16 for the beta activity. Gamma-ray spectra were obtained by combining and counting as one sample all the weekly samples collected each month from each location. Additional details on the sampling and counting techniques are given in ANL-5808 and ANL-5934.

TABLE II

LONG-LIVED ALPHA AND BETA ACTIVITIES IN WEEKLY AIR FILTER SAMPLES, 1960
(pc/m^3)

Month	Location	No. of Samples	Alpha Activity				Beta Activity			
			After 3 days		After 7 days		After 3 days		After 7 days	
			Max	Av	Max	Av	Max	Av	Max	Av
January	On-site	20	0.0072	0.0034	0.0047	0.0030	0.23	0.10	0.15	0.09
	Off-site	17	0.0078	0.0036	0.0053	0.0033	0.13	0.09	0.12	0.09
February	On-site	20	0.0063	0.0032	0.0054	0.0031	0.13	0.09	0.13	0.09
	Off-site	14	0.0059	0.0034	0.0058	0.0034	0.16	0.09	0.12	0.09
March	On-site	22	0.0090	0.0041	0.0090	0.0036	0.26	0.13	0.24	0.12
	Off-site	17	0.0052	0.0036	0.0054	0.0038	0.21	0.12	0.18	0.11
April	On-site	19	0.013	0.006	0.0055	0.0025	0.23	0.15	0.21	0.14
	Off-site	8	0.0066	0.0039	0.0038	0.0026	0.21	0.15	0.20	0.14
May	On-site	22	0.017	0.006	0.0045	0.0024	0.23	0.12	0.24	0.11
	Off-site	12	0.011	0.005	0.0045	0.0029	0.23	0.12	0.22	0.11
June	On-site	22	0.012	0.006	0.0046	0.0022	0.24	0.13	0.23	0.13
	Off-site	16	0.0079	0.0047	0.0032	0.0025	0.19	0.14	0.18	0.14
July	On-site	22	0.019	0.010	0.0045	0.0030	0.17	0.11	0.16	0.10
	Off-site	16	0.014	0.008	0.0058	0.0027	0.18	0.12	0.18	0.11
August	On-site	23	0.041	0.011	0.0054	0.0026	0.17	0.09	0.12	0.08
	Off-site	17	0.012	0.007	0.0046	0.0027	0.15	0.09	0.12	0.08
September	On-site	19	0.026	0.012	0.0054	0.0026	0.12	0.07	0.11	0.06
	Off-site	17	0.0093	0.0055	0.0046	0.0028	0.090	0.055	0.090	0.052
October	On-site	22	0.024	0.010	0.0060	0.0030	0.11	0.08	0.080	0.056
	Off-site	16	0.022	0.010	0.0057	0.0032	0.095	0.06	0.065	0.049
November	On-site	22	0.014	0.007	0.0064	0.0036	0.13	0.06	0.10	0.05
	Off-site	14	0.017	0.007	0.0059	0.0029	0.069	0.045	0.062	0.042
December	On-site	20	0.0097	0.0054	0.0072	0.0034	0.12	0.06	0.081	0.054
	Off-site	10	0.0093	0.0050	0.0045	0.0030	0.081	0.048	0.074	0.045
Annual Summary	On-site	253	0.041	0.007	0.0090	0.0029	0.26	0.10	0.24	0.09
	Off-site	174	0.022	0.006	0.0059	0.0030	0.23	0.09	0.22	0.09

TABLE III

LONG-LIVED ALPHA AND BETA ACTIVITIES IN WEEKLY AIR FILTER SAMPLES, 1961
(pc/m^3)

Month	Location	No. of Samples	Alpha Activity				Beta Activity			
			After 4 days		After 7 days		After 4 days		After 7 days	
			Max	Av	Max	Av	Max	Av	Max	Av
January	On-site	25	0.0059	0.0033	0.0044	0.0019	0.087	0.058	0.085	0.062
	Off-site	14	0.010	0.003	0.0058	0.0021	0.081	0.054	0.086	0.053
February	On-site	24	0.0064	0.0038	0.0046	0.0031	0.28	0.09	0.20	0.09
	Off-site	13	0.0067	0.0041	0.0059	0.0039	0.067	0.048	0.063	0.050
March	On-site	24	0.0056	0.0034	0.0048	0.0028	0.97	0.16	0.77	0.13
	Off-site	16	0.0045	0.0032	0.0045	0.0029	0.087	0.055	0.087	0.052
April	On-site	28	0.0067	0.0033	0.0059	0.0027	0.14	0.08	0.13	0.08
	Off-site	17	0.0060	0.0037	0.0052	0.0033	0.12	0.07	0.11	0.07
May	On-site	27	0.0061	0.0037	0.0046	0.0029	0.32	0.14	0.30	0.14
	Off-site	17	0.0063	0.0041	0.0059	0.0034	0.19	0.12	0.19	0.12
June	On-site	23	0.0069	0.0052	0.0054	0.0034	0.19	0.13	0.19	0.13
	Off-site	16	0.0067	0.0045	0.0059	0.0040	0.17	0.12	0.18	0.12
July	On-site	21	0.0088	0.0042	0.0042	0.0026	0.27	0.10	0.13	0.08
	Off-site	15	0.0073	0.0050	0.0057	0.0036	0.18	0.11	0.17	0.11
August	On-site	26	0.0094	0.0046	0.0064	0.0030	0.090	0.068	0.094	0.068
	Off-site	17	0.0068	0.0037	0.0044	0.0029	0.106	0.06	0.092	0.063
September	On-site	26	0.011	0.005	0.0056	0.0036	61.2	12.3	49.4	9.2
	Off-site	17	0.012	0.005	0.0067	0.0033	57.8	10.6	46.5	7.8
October	On-site	27	0.0085	0.0036	0.0060	0.0033	12.8	7.4	12.7	6.6
	Off-site	17	0.0062	0.0033	0.0057	0.0028	11.3	6.6	9.8	5.8
November	On-site	23	0.0061	0.0037	0.0050	0.0032	13.1	7.2	11.8	5.2
	Off-site	13	0.0058	0.0032	0.0044	0.0029	12.9	7.3	12.9	6.7
December	On-site	27	0.0065	0.0041	0.0068	0.0042	9.9	5.1	9.5	4.6
	Off-site	10	0.0054	0.0041	0.0062	0.0044	6.3	4.4	6.3	4.2
Annual Summary	On-site	301	0.011	0.004	0.0068	0.0031	61.2	2.7	49.4	2.2
	Off-site	182	0.012	0.004	0.0067	0.0033	57.8	2.5	46.5	2.1

The long-lived alpha and beta activities in the air samples collected during 1960 and 1961 are given in Tables II through V. The alpha activity remaining 3 or 4 days after collection was due primarily to natural activity present in dust in the air and to polonium-210 from the decay of radon-222. The average monthly or annual alpha activities were very similar both on and off the site, indicating that little, if any, of the airborne alpha activity in the environment was due to Argonne operations. The average differences between locations were less than the 10 to 20% error in sample collection and counting, although some individual samples differed from the average by a factor of up to about three. Such differences occurred at random both on and off the site and depend on a number of variables, such as the composition and amount of dust in the air at the time of sampling. The average seventh day alpha activities, approximately 0.003 pc/m^3 during 1960 and 1961, were very similar to those found earlier, as indicated in Table VI, and are considered normal. The effect of an additional day of decay on the alpha activity at 3 and 4 days is also apparent from Table VI.

TABLE IV

LONG-LIVED ALPHA AND BETA ACTIVITIES IN 24-HR AIR FILTER SAMPLES ON ANL SITE, 1960
(pc/m^3)

Month	No. of Samples	Alpha Activity*		Beta Activity*	
		Max	Av	Max	Av
January	27	0.018	0.005	0.24	0.07
February	25	0.011	0.003	0.22	0.07
March	27	0.021	0.006	0.29	0.12
April	28	0.037	0.015	0.33	0.14
May	31	0.060	0.020	0.25	0.12
June	29	0.073	0.021	0.33	0.15
July	31	0.078	0.025	0.22	0.12
August	31	0.086	0.024	0.22	0.10
September	25	0.068	0.022	0.22	0.09
October	29	0.086	0.040	0.38	0.13
November	30	0.052	0.018	0.17	0.08
December	30	0.029	0.011	0.10	0.07
Summary	343	0.086	0.018	0.38	0.11

*Activity remaining three days after end of sampling period.

TABLE V

LONG-LIVED ALPHA AND BETA ACTIVITIES IN 24-HR AIR FILTER SAMPLES ON ANL SITE, 1961
(pc/m^3)

Month	No. of Samples	Alpha Activity*		Beta Activity*	
		Max	Av	Max	Av
January	31	0.012	0.004	0.13	0.08
February	28	0.018	0.005	0.31	0.09
March	30	0.018	0.005	1.5	0.2
April	30	0.0094	0.0054	0.15	0.09
May	29	0.013	0.007	0.24	0.13
June	30	0.017	0.008	0.23	0.13
July	30	0.018	0.007	0.15	0.09
August	31	0.017	0.008	0.11	0.07
September	30	0.010	0.005	166.2	18.4
October	31	0.009	0.005	31.7	10.2
November	30	0.011	0.004	48.4	11.8
December	29	0.014	0.005	17.0	7.2
Summary	359	0.018	0.006	166	4.1

* Activity remaining four days after end of sampling period.

TABLE VI

AVERAGE ANNUAL RADIOACTIVITY IN AIR FILTER SAMPLES, 1953-1961
(pc/m^3)

Year	Location	Weekly Samples				Daily Samples 3-4 days	
		Alpha Activity		Beta Activity			
		3-4 days	7 days	3-4 days	7 days	Alpha Activity	Beta Activity
1953	On-site	0.010	0.006	1.2	1.0	0.03	1.4
1954	On-site	0.010	0.005	0.4	0.3	0.03	0.5
1955	On-site	0.007	0.004	0.8	0.7	0.03	1.0
1956	On-site	0.010	0.006	1.3	1.2	0.03	1.6
1957	On-site	0.008	0.005	2.1	1.7	0.021	2.0
	Off-site	0.007	0.004	2.1	1.7	-	-
1958	On-site	0.007	0.003	2.8	2.5	0.024	3.0
	Off-site	0.007	0.004	3.4	2.7	-	-
1959	On-site	0.006	0.0034	2.2	2.1	0.021	2.4
	Off-site	0.006	0.0039	2.3	2.2	-	-
1960	On-site	0.007	0.0029	0.10	0.09	0.018	0.11
	Off-site	0.006	0.0030	0.09	0.09	-	-
1961	On-site	0.004	0.0031	2.7	2.2	0.006	4.1
	Off-site	0.004	0.0033	2.5	2.1	-	-

In contrast with the relatively constant alpha activities found since air sampling was begun, the beta activities have varied widely due to varying amounts of fission product fallout. The long-lived beta activity during 1960, approximately 0.1 pc/m^3, was at the lowest level since sample collection was begun in 1953, since there was no large-scale nuclear testing in the atmosphere during 1959 and 1960. Fallout from the French nuclear detonations in the Sahara was not sufficiently large to affect the total beta activity in air. As will be seen, activity from this source was detected in a few precipitation samples. The beta activity was relatively constant from October 1959 through February 1960, being about 0.09 pc/m^3. From March through July 1960, there was a small but definite increase (to about 0.14 pc/m^3) in beta activity due to a "spring maximum" in stratospheric fallout. Fallout activity, as ascertained from the total beta activity, decreased slowly thereafter, and then remained fairly constant during the last quarter of 1960 and the first quarter of 1961. The increase in the on-site beta activity in February and March 1961 was due to small amounts of iodine-131 accidentally released at ANL and was not due to fallout. This release is discussed below.

A second "spring maximum" in fallout activity during May and June 1961 increased the beta activity to about the same level as the spring of 1960. Following this peak, the beta activity decreased until September 14, 1961, reaching about the same level as in December 1960. Beginning with the sample collected on September 14, 1961, and continuing through the remainder of the year, the beta activity was almost entirely due to fallout from the USSR series of nuclear tests that began on September 1, 1961. The sample collected on September 20, 1961, contained 166 pc/m^3 after 4 days, the highest beta activity thus far found in a single sample. The previous maximum was 127 pc/m^3 in May 1953.

The presence of short-lived fission products at all locations was confirmed by beta-decay rates, gamma-ray spectrometry, and chemical fission product analyses of both air and precipitation samples. The age of the fission products in the samples collected after September 14, 1961, ranged from 6 to 20 days. The similarity between the total beta activities, decay rates, and fission product concentrations on and off the site indicates that this beta activity was due to a widespread and relatively uniform source such as fallout and not to a localized source such as Argonne.

Concentrations of the principal particulate gamma-ray emitters in the air filter samples are given in Tables VII and VIII. The results listed under the ruthenium-rhodium pair of nuclides were obtained from the counting rate of gamma rays with energies in the neighborhood of 0.5 Mev after subtracting any activity due to barium-lanthanum-140. The principal nuclides in the air samples that emitted such gamma rays were beryllium-7, rhodium-102 and ruthenium-rhodium-106. The ruthenium and rhodium nuclides occur in fallout (ruthenium-rhodium-106 are fission products and rhodium-102 was added as a tracer in a nuclear test) and beryllium-7 is

TABLE VII

PRINCIPAL GAMMA-RAY ACTIVITY IN AIR FILTER SAMPLES, 1960
(pc/m^3)

Month	Cerium-144		Ruthenium-Rhodium-106*		Cesium-137	
	On-site	Off-site	On-site	Off-site	On-site	Off-site
January	0.013	0.013	0.010	0.015	0.008	0.004
February	0.020	0.012	0.024	0.018	0.005	0.004
March	0.018	0.017	0.028	0.022	0.007	0.007
April	0.033	0.033	0.036	0.043	0.011	0.012
May	0.025	0.024	0.034	0.037	0.009	0.010
June	0.027	0.027	0.050	0.053	0.011	0.011
July	0.033	0.043	0.044	0.053	0.016	0.021
August	0.021	0.026	0.063	0.071	0.016	0.021
September	0.014	0.013	0.041	0.042	0.0069	0.0067
October	0.012	0.0098	0.078	0.060	0.0068	0.0058
November	0.0078	0.0081	0.043	0.043	0.0047	0.0047
December	0.0090	0.0081	0.086	0.073	0.0063	0.0063
Average	0.019	0.020	0.045	0.044	0.0090	0.0095

* Gamma-ray activity with energies between 0.47 and 0.51 Mev, calculated as $Ru\text{-}Rh^{106}$ (see text).

TABLE VIII

PRINCIPAL GAMMA-RAY ACTIVITY IN AIR FILTER SAMPLES, 1961
(pc/m^3)

Month	Cerium-144		Ruthenium-Rhodium-106*		Cesium-137	
	On-site	Off-site	On-site	Off-site	On-site	Off-site
January	0.0082	0.0069	0.065	0.061	0.0073	0.0066
February	0.0085	0.0090	0.0071	0.066	0.0083	0.0081
March	0.0090	0.0093	0.059	0.062	0.0099	0.0093
April	0.0079	0.0069	0.059	0.051	0.0071	0.0056
May	0.017	0.014	0.088	0.068	0.015	0.011
June	0.015	0.014	0.078	0.084	0.014	0.011
July	0.011	0.014	0.053	0.070	0.0092	0.0117
August	0.0065	0.0058	0.048	0.043	0.0058	0.0051
September	0.21	0.20	0.096	0.082	0.0075	0.0062
October	0.23	0.30	0.11	0.11	0.0094	0.0080
November	0.31	0.35	0.16	0.16	0.0090	0.010
December	0.47	0.58	0.17	0.22	0.013	0.014
Average	0.11	0.13	0.083	0.085	0.096	0.0089
	Cerium-141		Zirconium-Niobium-95		Barium-Lanthanum-140	
September	0.53	0.33	0.91	0.82	7.5	6.4
October	0.85	0.74	1.13	1.08	3.2	2.7
November	0.69	0.77	1.33	1.42	1.9	1.7
December	0.54	0.76	1.50	1.64	0.58	0.67
September-December Average	0.65	0.65	1.2	1.3	3.3	2.9

* Gamma-ray activity with energies between 0.47 and 0.51 Mev, calculated as $Ru\text{-}Rh^{106}$ (see text).

produced in the upper atmosphere by cosmic rays. Thus, all three nuclides occur widely. Since the concentration of each of these nuclides in a complex mixture is difficult to determine by gamma-ray counting alone, the disintegration rate of this mixture was calculated as ruthenium-rhodium-106. Valid comparisons between the on-site and off-site samples may be made with results obtained in this way since the concentrations of all three nuclides are expected to be similar at all sampling locations in the absence of a localized release.

As shown in the tables, significant differences were found between the off-site and on-site samples for some of the nuclides during some months. However, these differences occurred at random in both directions, and were due to experimental error in resolving the complex gamma-ray spectra, errors in sample collection (primarily measurement of flow rate), and probably to small localized differences in the amount of fallout. The random differences in the monthly values and the small differences in the annual averages indicate that these gamma emitters originated in fallout or occurred naturally, and were not added to the air by Argonne. The increases in fallout rate during the spring, noted for the total beta activity, and the presence of short-lived fission products beginning in September 1961 are also evident in the gamma activity. The shorter-lived fission products, cerium-141, zirconium-niobium-95, and barium-lanthanum-140, were detected only after September 14, 1961. Concentrations of the longer-lived fission products, cerium-144, ruthenium-rhodium-106, and cesium-137, increased at the same time. There was little indication in the beta or gamma activity for a maximum in the fall of 1960 similar to that observed in the spring.

During February and March 1961, the beta activities on the ANL site were two to three times higher than those off-site. This increase on the site was due to iodine-131 removed from irradiated fuel elements by heating and accidentally released to the atmosphere in the exhaust air from two Argonne buildings. Iodine-131 produced in this way is generally considered to be gaseous and not particulate. However, 8% of the radioiodine was collected by the filter paper used. This collection efficiency was determined by field measurements, during this release, with charcoal traps in series with the filter paper and an assumption that the charcoal was completely efficient for the iodine. In experiments with several collection devices, including basic aqueous solutions and silver-loaded paper, charcoal proved to be the most efficient and reproducible iodine collector.

The iodine-131 concentrations found in the air samples during this period are given in Table IX. These values were obtained from gamma-ray spectra of the air filters, based on a collection efficiency of 8%, and were corrected for decay to the midpoint of the collection period, it being assumed that the iodine was added to the air at a constant rate during the collection.

TABLE IX

IODINE-131 IN AIR FILTER SAMPLES,
FEBRUARY-MARCH, 1961
(pc/m^3)

Month	Location	No. of Samples	Iodine-131 Concentration	
			Av	Max
February	On-site	20	1.9	15.4
	Off-site	14	<0.035	< 0.035
March	On-site	22	3.3	16.7
	Off-site	17	0.20	0.41

The iodine was first detected in some of the on-site air filters during the latter half of February, and was present during most of March at all sampling locations on the site. In March, it was also present at two of the four off-site locations (southwest and southeast of the site) at average concentrations of 0.41 and 0.36 pc/m^3 (0.14 and 0.12% of the MPC). The presence of iodine-131 is not apparent from the total beta activitiy in the off-site samples because both the concentration and collection efficiency were low. The maximum iodine-131 concentration on the site, 16.7 pc/m^3, is equivalent to 5.6% of the MPC; the average concentration on the site during February and March, 2.6 pc/m^3, amounted to 0.9% of the MPC. The beta activities in the daily air samples collected on the site (see Table V) also increased and varied with the iodine concentration during this period, and showed that the iodine was first noticeable in the air filter samples on February 17 and that the highest concentrations occurred from March 16 to 21.

The iodine release was essentially eliminated by the end of March. During the rest of the year only two on-site air samples contained detectable amounts of iodine-131 originating at Argonne. One sample, collected in April, contained 0.018 pc/m^3 (0.006% of the MPC) and one collected in May contained 0.034 pc/m^3 (0.011% of the MPC). None was detected in any of the off-site samples during this period.

Iodine-131 was, of course, detected in the air during the latter part of 1961. However, the concentrations were relatively low compared with the other short-lived fission products, and were essentially the same both on and off the site. This iodine apparently resulted from the USSR nuclear test series; there was no evidence to indicate that it originated at Argonne. The iodine released by Argonne was also detected in soil, grass, surface water, and rain on the ANL site. These results are given in the appropriate sections of this report.

B. Precipitation

Precipitation was collected on the ANL site at location 8F in Figure 13. The rain collector was arranged to collect two successive one-gallon portions when the rainfall was sufficiently large, each portion being equivalent to 0.48 in. of rain. A collector of this size is sufficient to collect most of the rain during the year and still provide samples sufficiently large for analysis from rainfalls totaling only a few hundreths of an inch or from rainfalls containing little activity. Snow was also collected from the rain tray whenever possible in order to obtain a sample from a known area. When two portions were collected, they were analyzed as separate samples.

The total alpha and beta activities during 1960 and 1961 are given in Tables X and XI. The alpha activities were similar to those normally

TABLE X

NONVOLATILE RADIOACTIVITY IN PRECIPITATION AT ANL, 1960

Month	No. of Samples	Time (days)*	Alpha Activity (pc/liter)		Beta Activity (pc/liter)		Beta Activity Total (mc/sq mi)
			Max	Av	Max	Av	
January	3	1	1.9	1.8	52	35	-
	3	7	1.7	0.9	51	33	2.7
February	2	1	6.9	4.1	74	6	-
	4	7	5.1	3.5	434	151	10.0
March	4	1	14.1	5.0	350	112	-
	3	7	8.8	3.8	340	127	13.9
April	7	1	41.9	10.8	220	96	-
	7	7	3.5	2.0	173	83	11.0
May	6	1	9.7	4.2	95	56	-
	6	7	1.0	0.8	84	49	8.1
June	9	1	17.9	6.0	275	85	-
	9	7	6.4	1.5	246	71	6.7
July	4	1	12.8	5.2	56	34	-
	6	7	0.9	0.6	35	22	3.3
August	4	1	11.2	7.5	56	44	-
	5	7	0.7	0.5	35	26	2.7
September	6	1	42.0	10.0	237	58	-
	6	7	15.0	2.9	183	42	1.5
October	4	1	16.6	8.3	96	52	-
	5	7	4.9	1.8	49	24	2.4
November	6	1	8.5	3.7	47	23	-
	6	7	4.3	1.2	41	16	1.3
December	2	1	1.9	1.9	18	15	-
	2	7	1.8	1.2	17	11	0.3
Annual Summary	57	1	42.0	6.3	350	59	-
	62	7	15.0	1.7	434	54	63.9

*Time after end of precipitation (approximately).

TABLE XI

ALPHA AND BETA ACTIVITIES IN PRECIPITATION AT ANL, 1961

Month	No. of Samples	Time[1] (days)	Alpha Activity (pc/liter)		Beta Activity (pc/liter) x 10^{-2}		Total (mc/sq mi)	Age Range[2] (days)
			Max	Av	Max	Av		
January	1	1	3.4	3.4	2.05	2.05	-	-
	2	7	7.2	5.2	2.78	2.46	1.6	-
February	2	1	2.9	1.6	0.15	0.09	-	-
	2	7	0.9	0.6	0.15	0.09	0.1	-
March	10	1	53	11	1.92	0.71	-	-
	10	7	6.1	2.3	1.63	0.57	16.3	-
April	7	1	17	5	1.09	0.61	-	-
	7	7	3.3	1.9	0.66	0.44	6.4	-
May	6	1	23	11	1.18	0.67	-	-
	6	7	4.6	7.2	0.80	0.55	5.6	-
June	7	1	21	8	0.88	0.50	-	-
	7	7	2.6	1.4	0.75	0.41	4.5	-
July	8	1	35	6	1.10	0.28	-	-
	8	7	4.2	1.0	0.58	0.17	3.2	-
August	7	1	12	4	0.37	0.23	-	-
	9	7	3.3	1.3	0.29	0.19	3.4	-
September	14	1	17	4	70.1	6.50	-	-
	16	7	1.7	0.7	56.9	4.90	75.7	15->300
October	8	1	6.7	4.1	51.6	19.7	-	-
	8	7	5.1	1.5	41.0	15.9	305	18-37
November	8	1	23	6	52.8	29.7	-	-
	8	7	1.8	0.8	50.8	25.0	289	17-50
December	5	1	7.5	3.2	38.2	26.8	-	-
	5	7	2.4	1.6	33.7	22.7	241	15-50
Annual	83	1	53	6	70.1	7.8	-	-
Summary	88	7	7.2	1.8	56.9	6.2	952	-

[1] Days after end of precipitation (approximately).

[2] Calculated assuming a beta-decay rate proportional to $T^{-1.2}$.

found in rain during other years. In 1959, for example, the monthly average alpha activities varied from 0.3 to 13.8 pc/liter and averaged 2.1 pc/liter after 7 days of decay. The alpha activity was due primarily to naturally occurring radioactive nuclides present in the air.

The beta activity was due primarily to fallout from nuclear detonations. The monthly variations in beta activity can be correlated approximately with the beta activity in the air filter samples, although quantitative

correspondence cannot be expected since the air filter samples were collected continuously and precipitation is an intermittant sampler of the atmospheric radioactivity. Precipitation, however, is a more sensitive indicator of airborne activity, since it samples a much larger volume of air than the air filters. The beta activities in the air filter and precipitation samples are compared in Figure 15. The spring maxima observed in the air filter activities are not as obvious in the precipitation results, and the fluctuations in precipitation activities were more pronounced and irregular than in air.

Fallout from the French nuclear tests conducted in the Sahara in February 1960 was detected in precipitation during February and March 1960. A sample collected on February 22, 1960 contained 430 pc/liter of beta activity, about 10 times the average activity during the previous 5 months. Short-lived fission products (barium-140 and strontium-89) were detected in this sample, and the beta-decay rate indicated a detonation date of February 12, 1960. The bulk of the fission products in this sample evidently originated in the test on February 13, 1960. A rainfall on March 9 also contained short-lived fission products.

The other samples collected during 1960 contained little, if any, fallout from the French tests, as shown by the beta decay rate. Notably, a sample collected on March 30 contained an abnormal amount of beta activity, 340 pc/liter after 7 days. Short-lived fission products were not found in this sample, and the decay rate indicated an age greater than 1 year. There was no precipitation between February 22 and March 6, and it is likely that additional fallout from the French tests would have been present in precipitation during this period.

Fission products from the French tests were not detected in the air filter samples. Apparently, the concentration in surface air was too low, and the fallout was removed from the air at higher altitudes by the precipitation.

From February 1960 through December 1960, the beta activity in precipitation generally decreased. Occasional samples contained greater than 100 pc/liter (two in June and one in September), and these few samples caused peaks in the monthly averages during this period. The beta-decay rates during this period indicated that the age of the fission products in these samples was greater than 1 year.* The beta activity increased sharply in January 1961; the two samples during this month each contained more than 200 pc/liter. Both samples, however, contained only long-lived

*The ages cannot be calculated from the first and seventh day activities given in the tables since radon and thoron decay products were present in some of the samples at the time of the first count. Other counts not given in the tables were used for age calculations.

fission products. The average beta activity increased again in March, primarily because of iodine-131 resulting from the release discussed in Section III-A.

On-site surface water also contained iodine-131 washed out of the air by rainfall during this period. After March, the beta activity showed the maximum observed in the air filter samples and, beginning in September, the effect of fallout from the USSR test series.

The total amount of beta activity carried down by precipitation, as determined from the seventh day activity, was only 64 mc/mi^2 in 1960. During 1961, the total deposition of beta activity increased to 952 mc/mi^2, and 95% of this activity resulted from the nuclear testing beginning in September. During previous years the total beta deposition varied from about 600 mc/mi^2 in 1953, 1954, and 1956 to 11,000 mc/mi^2 in 1952. It is interesting to note that the alpha activities during 1960 and 1961 showed the same general trends as the beta activities except for the sharp increase in beta activity from September to December 1961. In the absence of tropospheric fallout, apparently many of the same variables influence both the alpha and beta activities.

Fission product activity in selected precipitation samples collected after the resumption of atmospheric testing in September 1961 is shown in Table XII. For comparison purposes, the concentrations of strontium-90 and cesium-137 prior to the appearance of fallout from the 1961 test series were 0.2-0.3 pc/liter, and the shorter-lived iodine-131, barium-140, and strontium-89 could not be detected. Based on ratios of the strontium-89

TABLE XII

FISSION PRODUCT ACTIVITY IN SELECTED PRECIPITATION SAMPLES, SEPTEMBER-DECEMBER, 1961
(pc/liter)

Date of Precipitation	Amount of Precipitation (in.)	Total Beta Activity (x 10^2)	I^{131} (x 10^2)	Ba^{140} (x 10^2)	Sr^{89} (x 10^2)	Sr^{90}	Cs^{137}
September 23-25 (first portion)	3.86	5.7	1.3	1.5	0.5	0.5	4.3
October 13	0.40	15.9	0.97	2.9	1.8	1.5	2.7
October 20 (first portion)	1.03	17.3	1.4	4.0	1.9	1.5	1.4
October 27-29	0.86						
(first portion)		41.0	-	5.6	2.7	2.7	3.9
(second portion)		16.5	-	4.5	2.0	2.3	-
November 2-3	0.76						
(first portion)		30.5	4.2	9.2	5.6	4.7	2.9
(second portion)		31.8	-	12.1	6.7	5.4	-
November 13	0.10	30.2	1.5	11.1	10.4	11.2	-
November 16	0.48	10.6	-	1.9	1.6	1.6	-
December 16	0.11	18.8	-	2.2	4.8	6.3	-
December 22-23	1.06	28.2	0.10	1.2	3.6	6.2	-

to strontium-90 and of the barium-140 to strontium-89 concentrations in the table, the ages of the fission products in the precipitation from September 23 through November 16 varied from about 20 to 45 days. In the December precipitation the age appeared to be about 80 days, indicating the presence of fission products produced, on the average, late in September, although a preponderance of fission products from the multimegaton tests conducted on October 23 and 30 might have been expected. Fallout from the latter tests appeared to be present in the air during the middle of November. However, all samples probably contained fission products from a number of detonations carried out at different times, so that only average fission product ages can be calculated from the data.

C. Water

1. Sawmill Creek and Des Plaines River

Argonne waste water is discharged into Sawmill Creek at location 7M in Figure 13, and this stream was sampled before it passed any of the ANL buildings (location 13L) and below the waste water outfall (location 7M) to determine if radioactive materials were added to the stream in the waste water. Samples were collected below the outfall three times weekly. During 1960 each below-outfall sample was analyzed individually for some or all of the nuclides of interest. In 1961 equal portions of each of the three below-outfall samples collected each week were combined prior to analysis. In this way the average activity in the three samples was obtained. Since the activity in the below-outfall samples generally varied widely from day to day, a representative average activity could be obtained only if all the samples were analyzed separately, or if the samples were combined prior to analysis. The latter practice was adopted in 1961 since it was impractical to analyze all the samples for all nuclides of interest.

Above-site samples were collected at weekly intervals, and at least one sample each month was analyzed for each nuclide of interest. When the stream contains little fallout activity, the radioactivity above the outfall consists primarily of natural activities and remains fairly constant with time, and the average activity during any period is close to the activity found in a few samples collected during the same period. When the amount of fallout is low, the results obtained from such a sampling and analytical schedule can be compared directly to determine the Argonne contribution to the below-outfall water.

From the middle of September 1961 to the end of the year, when fallout activity in the creek varied greatly and contributed a significant share of the total beta activity, the number of fission product analyses performed on above-site water was increased to obtain a better value for the average beta activity at that location. However, during such periods direct

comparisons of above- and below-outfall beta activities cannot solely be used to evaluate the Argonne contribution to the stream, since fallout is added to the stream below the outfall directly from the air (primarily by precipitation) as well as by above-site water. Under these conditions it is difficult to evaluate the Argonne contribution accurately, and additional evidence, such as detonation dates and air and precipitation activities, are useful.

The alpha activities found in Sawmill Creek water during 1960 and 1961 are given in Tables XIII and XIV. The average alpha activity above the outfall, approximately 1.9 pc/liter during both years, was due primarily to radioactive nuclides that occur naturally in the stream. The additional alpha activity below the outfall was evidently added to the stream in Argonne waste water. Since creek water is diluted on the average approximately in half by Argonne waste water, the average alpha activity below the outfall due to natural creek water was about 0.9 pc/liter, or one-half of the above-site concentration. The additional activity below the outfall, about 29 pc/liter in 1960 and 6 pc/liter in 1961, resulted from Argonne waste water.

TABLE XIII

NONVOLATILE ALPHA ACTIVITY IN SAWMILL CREEK WATER, 1960

Month	Location[1]	Total Alpha Activity (pc/liter)			Uranium (pc/liter)			Plutonium (pc/liter)			Thorium (pc/liter)		
		No. of Samples	Max	Av	No. of Samples	Max	Av	No. of Samples	Max	Av[2]	No. of Samples	Max	Av[2]
January	13L	3	6.1	3.1	3	3.5	2.0	1	<0.05	-	1	0.23	-
	7M	12	366	81.7	7	448	156	2	<0.05	-	2	<0.05	-
February	13L	2	2.4	2.0	2	2.0	1.7	1	<0.05	-	1	<0.05	-
	7M	12	850	132	7	863	209	4	0.50	0.20	4	0.21	0.07
March	13L	3	3.2	2.7	3	1.8	1.3	1	<0.05	-	1	<0.05	-
	7M	11	61.6	20.2	7	54.3	22.4	5	0.54	0.16	4	<0.05	-
April	13L	4	3.2	2.3	4	1.4	1.1	3	<0.05	-	3	0.26	0.10
	7M	13	49.0	16.1	7	37.2	16.1	6	0.27	0.13	6	0.05	0.03
May	13L	4	2.1	1.5	4	1.8	1.4	2	<0.05	-	2	<0.05	-
	7M	12	74.9	22.3	8	81.9	32.1	3	0.08	0.04	3	<0.05	-
June	13L	5	2.5	2.1	5	1.6	1.4	2	<0.05	-	2	<0.05	-
	7M	13	233	35	7	249	64	4	0.15	0.06	4	1.1	0.44
July	13L	4	2.7	2.0	4	2.0	1.5	2	<0.05	-	2	<0.05	-
	7M	11	55.1	9.3	7	50.3	12.5	4	0.13	0.06	4	0.26	0.13
August	13L	5	1.3	1.1	5	0.8	0.7	2	<0.05	-	2	<0.05	-
	7M	14	15.1	6.6	9	19.2	7.0	5	0.48	0.14	5	0.50	0.17
September	13L	4	4.9	1.8	4	0.7	0.6	3	<0.05	-	3	<0.05	-
	7M	12	45.6	10.4	6	17.5	7.3	5	0.09	0.06	5	1.5	0.60
October	13L	4	1.4	1.0	4	0.9	0.7	2	<0.05	-	2	<0.05	-
	7M	13	20.6	7.0	6	18.9	8.0	3	0.11	0.08	3	0.69	0.34
November	13L	5	2.4	1.7	5	1.8	1.1	2	<0.05	-	2	<0.05	-
	7M	12	13.3	7.5	6	9.8	4.6	3	<0.05	-	3	<0.05	-
December	13L	3	1.9	1.4	2	1.5	1.0	1	<0.05	-	1	<0.05	-
	7M	11	24.1	8.1	6	20.0	7.1	3	0.06	0.04	3	0.45	0.18
Annual Summary	13L	46	6.1	1.8	45	3.5	1.2	22	<0.05	<0.05	22	0.26	0.05
	7M	146	850	30	83	863	46	47	0.54	0.09	47	1.1	0.18

[1]Location 13L is upstream from the ANL site. Location 7M is downstream from the waste water outfall. See Figure 13.

[2]Averages were calculated assuming a concentration of 0.025 pc/liter, one-half of the minimum detectable concentration, for those samples in which plutonium or thorium were not detected.

TABLE XIV

NONVOLATILE ALPHA ACTIVITY IN SAWMILL CREEK WATER, 1961

Month	Location[1]	Total Alpha Activity (pc/liter)			Uranium (pc/liter)			Plutonium (pc/liter)			Thorium (pc/liter)		
		No. of Samples	Max	Av	No. of Samples	Max	Av	No. of Samples	Max	Av[2]	No. of Samples	Max	Av[2]
January	13L	4	1.4	0.9	2	0.54	0.47	1	<0.05	-	1	0.08	-
	7M	12	13.8	9.7	12	10.5	8.4	12	0.20	0.10	12	0.25	0.11
February	13L	4	3.9	2.1	2	1.9	1.2	1	<0.05	-	1	<0.05	-
	7M	12	11.9	9.0	12	10.6	8.1	12	0.24	0.13	12	0.13	0.06
March	13L	5	5.5	3.7	3	2.0	1.9	1	<0.05	-	1	<0.05	-
	7M	15	7.4	5.1	15	4.9	3.5	12	0.06	0.04	12	<0.05	-
April	13L	4	2.6	2.1	2	1.5	1.4	1	<0.05	-	1	<0.05	-
	7M	12	5.9	4.2	12	5.1	3.4	12	0.05	0.04	12	<0.05	-
May	13L	5	1.7	1.4	2	1.6	1.4	1	<0.05	-	1	<0.05	-
	7M	12	5.4	3.4	12	2.9	2.4	12	0.21	0.09	12	0.25	0.10
June	13L	4	4.0	2.6	2	1.4	1.2	1	<0.05	-	1	<0.05	-
	7M	15	64.1	18.4	15	52.1	14.8	14	0.11	0.06	14	0.25	0.05
July	13L	4	1.7	1.4	2	1.0	1.0	1	<0.05	-	1	<0.05	-
	7M	12	8.1	5.2	12	6.0	4.4	12	0.70	0.33	12	<0.05	-
August	13L	5	3.4	1.9	2	1.4	1.3	1	<0.05	-	1	<0.05	-
	7M	15	17.4	7.7	15	16.9	8.0	14	<0.05	-	14	0.08	0.04
September	13L	4	1.6	1.2	2	1.7	1.5	1	<0.05	-	1	<0.05	-
	7M	9	7.1	5.7	9	7.1	4.8	9	0.10	0.06	9	<0.05	-
October	13L	4	1.8	1.4	2	0.93	0.92	1	<0.05	-	1	<0.05	-
	7M	15	3.9	2.9	15	3.0	2.1	15	<0.05	-	15	0.08	0.04
November	13L	5	2.2	1.8	2	1.1	1.0	1	<0.05	-	1	<0.05	-
	7M	12	8.1	4.0	12	6.4	2.8	12	0.17	0.06	12	<0.05	-
December	13L	4	3.1	2.6	2	1.3	1.2	1	<0.05	-	1	<0.05	-
	7M	15	10.6	6.4	15	9.1	4.8	14	0.27	0.05	14	0.58	0.08
Annual Summary	13L	52	5.5	1.9	25	2.0	1.2	12	<0.05	-	12	0.08	0.03
	7M	156	64.1	7.0	156	52.1	5.7	153	0.70	0.08	153	0.58	0.05

[1]Location 13L is upstream from the ANL site. Location 7M is downstream from the waste water outfall. See Figure 13.

[2]Averages were calculated assuming a concentration of 0.025 pc/liter, one-half of the minimum detectable concentration, for those samples in which plutonium or thorium were not detected.

A comparison of the total alpha activities and uranium concentrations shows that most of the activity added by Argonne was due to normal uranium. The uranium concentration added to the creek in Argonne waste water, assuming a dilution of one-half below the outfall, was about 45 pc/liter in 1960 and 5 pc/liter in 1961. These concentrations are equivalent to only 0.11 and 0.013% of the MPC, respectively.* Since some of the 1960 below-outfall samples that contained below average amounts of total alpha activity were not analyzed for uranium, while all samples were analyzed for total alpha activity, the average uranium concentrations (46 pc/liter) was greater than the average total alpha activity (30 pc/liter). If the average total alpha activity is taken as the average uranium concentration

*The MPC given by the ICRP for uranium and thorium is based upon a special definition of the curie such that one "curie" of recently extracted uranium or thorium is actually equivalent to (very nearly) 2c of uranium (U^{238} + U^{234} + U^{235}) and thorium (Th^{232} + Th^{228}). Thus, the MPC given by the ICRP must be multiplied by two for comparison with the concentrations given in this report, since these concentrations were obtained with use of the conventional definition of the curie.

in all below-outfall samples collected (this is very nearly correct since a comparison of individual samples shows that uranium accounts for essentially all the alpha activity in samples analyzed for both total alpha activity and uranium and since significant concentrations of other alpha emitters were not detected), the average uranium concentration contributed by the waste water was approximately 29 pc/liter, or 0.073% of the MPC. In previous years the average annual alpha activity varied from 9.6 to 33 pc/liter, and in each year most of the alpha activity was due to uranium. Thus, the alpha activity (and uranium concentration) in 1960 was about the same as the highest average previously found, whereas in 1961 it was the lowest thus far obtained.

Exact correspondence between the total alpha and uranium activities cannot be expected even when uranium is the only alpha emitter present, because of the errors inherent in determining the total alpha activity by counting thick samples and because the uranium isotopic composition is unknown. The uranium concentration was determined fluorometrically (because of its speed and accuracy for small amounts of natural or depleted uranium) and converted to activity through use of the specific activity of natural uranium, the most commonly encountered isotopic mixture. However, a variety of isotopic mixtures is used at Argonne, and the alpha activity due to uranium in a given sample can be larger or smaller than the activity calculated from the fluorometric analysis if the sample contains enriched or depleted uranium. It was impractical to determine for each sample the isotopic composition or the uranium activity by separation and counting. However, samples in which large differences were found between the total alpha and uranium activities received additional study to determine the true uranium content. In 1959 a large number of such samples were found, but in 1960 and 1961 this problem was encountered only rarely, and the effect of these samples on the average uranium concentration was small.

Small amounts of plutonium and thorium were also found in about one-half of the below-outfall samples as a result of their presence in Argonne waste water. Plutonium in concentrations greater than 0.05 pc/liter was not detected above the site and thorium was found in greater average concentrations below the outfall than above. The average Argonne contributions during both years, from the values given in the tables, amounted to only 0.0017 and 0.010% of the MPC for plutonium and thorium, respectively. If the average concentrations are calculated based on an assumption that there existed a concentration of zero instead of 0.025 pc/liter in those samples containing less than the minimum detectable concentrations, the average plutonium and thorium concentrations are reduced by 15 and 40%, respectively. The isotopic composition of the thorium was not specifically investigated, but the growth of alpha activity in the separated thorium fractions indicated the presence of equal activities of thorium-232 and thorium-228.

The distribution of alpha activity in Sawmill Creek during 1960 and 1961 followed the same pattern found in previous years. Below the outfall the creek has usually contained some uranium resulting from Argonne waste water. Plutonium, thorium, and other alpha emitters have been found much less frequently and in much smaller concentrations.

The total beta activities and the concentrations of the most frequently encountered beta emitters, except thorium-234 (UX_1), are given in Tables XV and XVI. The beta activity above the site prior to September 15, 1961, usually ranged between 5 and 10 pc/liter, although a few samples were as high as 15 pc/liter. This is the normal range at this location when the fallout activity is small compared with the natural beta activity. The additional activity below the outfall prior to September 15, 1961, can be attributed to contamination in Argonne waste water, and the Argonne contribution, principally due to strontium-90, cesium-137, cobalt-58, cobalt-60, and thorium-234, can be calculated as was done for the alpha activity. A method for estimating thorium-234 is given below; concentrations of the other nuclides are given in the tables. However, for the fission products, strontium-90 and cesium-137, the total concentrations, regardless of source (fallout or Argonne), must be used for comparisons with the MPC's since these nuclides are not naturally present in the environment.

During the last quarter of 1961, when fallout from USSR tests contributed most of the beta activity in the creek, the amount of fission products originating in Argonne waste water was more difficult to determine. By comparing results for individual samples from all locations collected at about the same time, from the ratios of fission products of different half-lives and from the fallout activity in precipitation, it is possible to draw the following conclusions. The strontium-89, strontium-90, barium-140, and iodine-131 in the below-outfall samples during this period resulted primarily from fallout, while cesium-137 was added by both Argonne waste water and fallout. Concentrations of cesium-137 in excess of 1.5 to 2 pc/liter may be attributed entirely to Argonne waste water. For example, the below-site sample containing the largest concentration of beta activity during this period, 91 pc/liter, was collected on November 1, 3, and 6. The barium-140/strontium-89, strontium-89/strontium-90, and strontium-90/cesium-137 ratios in this sample were approximately the same as those in an above-site sample collected on November 1, although the above-site sample contained about 3.5 times less activity. However, a heavy rainfall, totaling 0.76 in. and containing about 3000 pc/liter of total beta activity, occurred on November 2 and 3, and the increased total activity below the site can be attributed to this source.

TABLE XV

BETA ACTIVITY IN SAWMILL CREEK WATER, 1960

Month	Location[1]	Total Beta Activity (pc/liter)			Strontium-90 (pc/liter)			Cobalt-58 (pc/liter)			Cobalt-60 (pc/liter)			Cesium-137 (pc/liter)		
		No. of Samples	Max	Av	No. of Samples	Max	Av[2]	No. of Samples	Max	Av[2]	No. of Samples	Max	Av[2]	No. of Samples	Max	Av[2]
January	13L	3	15.0	9.3	2	1.3	1.2	1	<5	-	1	<2	-	1	<0.5	-
	7M	12	120	36	5	1.4	1.1	2	884	495	2	<2	-	3	17	7
February	13L	2	4.5	4.5	1	1.1	-	1	<5	-	1	<2	-	0	-	-
	7M	10	201	47	2	0.5	0.5	3	2500	850	3	171	87	1	1.2	-
March	13L	3	10.1	6.8	2	1.3	0.9	1	<5	-	1	<2	-	1	<0.5	-
	7M	11	155	65	4	3.4	1.5	3	1970	855	3	22	14	2	10.3	5.7
April	13L	4	8.2	6.0	2	1.6	1.2	1	<5	-	1	<2	-	3	0.9	0.4
	7M	13	47.1	28.7	7	3.7	3.2	4	344	262	4	64	20	8	6.2	2.1
May	13L	4	5.4	4.3	2	2.0	1.1	1	<5	-	1	<2	-	2	0.5	0.5
	7M	12	79.9	26.8	7	2.0	1.3	3	196	105	3	7.7	4.5	6	4.7	2.7
June	13L	5	5.3	4.4	2	1.0	0.8	1	<5	-	1	<2	-	1	<0.5	-
	7M	13	177	32.8	5	2.0	1.5	3	540	185	3	<2	-	4	4.2	2.0
July	13L	4	9.1	6.7	2	0.8	0.7	2	<5	-	2	<2	-	2	0.6	0.5
	7M	11	424	57	4	7.0	3.5	6	2650	815	6	8.5	3.9	5	83	26
August	13L	5	6.7	5.6	2	<0.5	-	1	<5	-	1	<2	-	1	<0.5	-
	7M	14	158	41	7	4.6	2.2	3	1580	655	3	130	49	7	18.5	8.7
September	13L	4	5.8	4.4	2	0.8	0.6	1	<5	-	1	<2.	-	1	<0.5	-
	7M	12	76.1	26.3	4	7.1	3.1	4	47.8	28.3	4	2.5	1.7	6	11.3	5.1
October	13L	4	5.6	4.8	2	1.0	0.6	1	<5	-	1	<2	-	1	<0.5	-
	7M	13	35.5	17.1	5	1.5	0.8	3	<5	-	3	<2	-	3	8.6	5.9
November	13L	5	10.8	7.0	2	0.8	0.5	1	<5	-	1	<2	-	1	<0.5	-
	7M	12	32.1	15.9	6	2.6	1.6	3	68	45	3	21	11	2	5.3	3.4
December	13L	3	5.8	5.5	2	<0.5	-	1	<5	-	1	<2	-	2	<0.5	-
	7M	11	472	66.4	6	182	32	3	<5	-	3	<2	-	3	8.3	5.3
Annual Summary	13L	46	15.0	5.8	23	2.0	0.8	13	<5	-	13	<2	-	16	0.9	0.3
	7M	144	424	37	62	182	5	40	2650	369	40	171	13	50	83	7

[1]Location 13L is upstream from the ANL site. Location 7M is downstream from the waste water outfall. See Figure 13.

[2]Averages were calculated assuming a concentration of one-half of the minimum detectable concentration for those samples in which a given nuclide was not detected.

TABLE XVI

BETA ACTIVITY IN SAWMILL CREEK WATER, 1961

Month	Location[1]	Total Beta Activity (pc/liter)			Strontium-90 (pc/liter)			Strontium-89 (pc/liter)			Cobalt-58 (pc/liter)			Cobalt-60 (pc/liter)			Cesium-137 (pc/liter)			Barium-140 (pc/liter)			Iodine-131 (pc/liter)		
		No. of Samples	Max	Av	No. of Samples	Max	Av[2]	No. of Samples	Max	Av[2]	No. of Samples	Max	Av[2]	No. of Samples	Max	Av[2]	No. of Samples	Max	Av[2]	No. of Samples	Max	Av[2]	No. of Samples	Max	Av[2]
January	13L	4	5.4	4.7	1	<0.5	-	1	<1	-	0	-	-	0	-	-	1	<0.5	-	1	<2	-	2	<2	-
	7M	12	23.2	19.5	12	2.6	1.8	12	<1	-	3	<5	-	3	1.2	-	12	3.7	2.8	9	5.6	2.9	12	<2	-
February	13L	4	8.3	6.6	1	<0.5	-	1	<1	-	1	<5	-	1	<2	-	1	<0.5	-	1	<2	-	0	-	-
	7M	12	29.8	26.5	12	1.5	1.2	12	<1	-	6	<5	-	6	1.3	1.0	12	3.2	1.8	1	<2	-	12	2	1
March	13L	5	14.3	8.9	1	0.65	-	1	<1	-	1	<5	-	1	<2	-	1	<0.5	-	1	<2	-	2	2.5	1.8
	7M	15	33.5	16.6	15	4.1	1.8	15	<1	-	6	6.7	4.6	6	<2	-	15	1.6	0.8	15	<2	-	15	55	26
April	13L	4	6.8	6.0	1	<0.5	-	1	<1	-	1	<5		1	<2	-	1	<0.5	-	1	<2	-	1	<2	-
	7M	12	22.6	15.8	12	1.0	0.8	12	1.6	1.0	6	9.0	7.5	6	<2	-	12	11.3	3.5	12	<2	-	12	2.4	1.8
May	13L	5	8.7	7.3	1	0.70	-	1	<1.	-	1	<5	-	1	<2	-	1	<0.5	-	1	<2	-	0	-	-
	7M	12	22.2	17.6	12	1.9	0.9	12	2.0	0.9	6	39	36	6	2.0	1.3	12	3.8	2.4	12	<2	-	9	<2	-
June	13L	4	8.3	6.5	1	<0.5	-	1	<1	-	1	<5	-	1	<2	-	1	<0.5	-	1	<2	-	1	<2	-
	7M	15	59.6	38.6	15	4.6	2.3	15	<1	-	15	231	88	15	8.8	5.1	15	4.1	2.3	15	<2	-	15	3	1.4
July	13L	4	8.1	6.8	1	0.91	-	1	<1	-	1	<5	-	1	<2	-	1	0.5	-	1	<2	-	1	<2	-
	7M	12	127	43.7	12	0.7	0.5	12	<1	-	12	735	202	12	26	8	12	2.3	1.7	12	<2	-	12	<2	-
August	13L	5	7.4	5.8	1	1.2	-	1	<1	-	1	<5	-	1	<2	-	1	<0.5	-	0	<2	-	0	<2	-
	7M	15	72.5	36.3	15	9.8	3.0	15	<1	-	15	101	46	15	1.6	1.1	15	11.0	3.6	15	<2	-	3	<2	-
September	13L	4	14.7	10.4	2	1.1	-	2	<1	-	1	<5	-	1	<2	-	1	1.0	-	1	2.5	-	0	<2	-
	7M	9	45.7	29.6	9	1.1	1.0	9	3.7	1.6	9	50	2.3	9	218	79	9	1.8	1.0	9	2.5	1.6	0	<2	-
October	13L	4	19.1	13.1	2	0.65	-	2	4.4	-	1	<5	-	1	<2	-	2	1.0	-	2	4.2	3.4	2	5	4
	7M	15	70.7	33.6	15	1.1	0.9	15	8.0	5.4	15	616	130	15	13	3.7	15	3.2	1.6	15	6.4	4.2	15	8	4
November	13L	5	34.1	26.5	2	0.80	-	2	8.1	-	1	<5	-	1	<2	-	2	1.0	-	2	4.2	3.5	2	9	8
	7M	12	90.5	50.9	12	1.5	1.1	12	29.2	17.4	12	52	20	12	1.5	0.9	12	2.1	1.0	12	11	7	12	11	5
December	13L	4	29.8	22.4	4	1.0	0.7	4	12.7	9.9	1	<5	-	1	<2	-	2	<0.5	-	1	<2	-	1	<2	-
	7M	15	55.3	36.8	15	1.1	0.7	15	7.9	6.6	15	13.4	6.3	15	2.3	1.6	15	6.5	2.8	15	5.0	2.4	15	<2	-
Annual Summary	13L	52	34.1	10.6	18	1.2	0.6	18	12.7	2.2	11	<5	-	11	<2	-	15	1.0	0.5	13	4.2	1.8	12	9	2.8
	7M	156	127	30.7	156	9.8	1.4	156	29.2	3.0	120	735	59	120	218	8	156	11.3	2.1	153	11	2	132	55	5

[1]Location 13L is upstream from the ANL site. Location 7M is downstream from the waste water outfall. See Figure 13.

[2]Averages were calculated assuming a concentration of one-half of the minimum detectable concentration for those samples in which a given nuclide was not detected.

The Argonne contribution of the cobalt nuclides and uranium daughters (thorium-234 and protactinium-234) during periods of fallout can be readily obtained as before, since fallout contains little, if any, of these nuclides. The uranium-daughter contribution can be estimated from the uranium concentrations and total alpha activities given in Tables XIII and XIV as follows. The measured beta-disintegration rate (as thallium-204, the nuclide on which the total beta analysis is based) due to thorium-234 and protactinium-234 in equilibrium with the parent uranium is equivalent to about 90% of the uranium concentration with the type of sample preparation, counting, and self-absorption and other correction factors used in obtaining total beta activities in water. Analyses for thorium-234 in some of the below-outfall samples indicated that about 70% of the equilibrium amount was present on the average in the water. Thus, the average thorium-234 and protactinium-234 activities are equal to approximately 60% of the uranium activity.

The average and maximum concentrations of beta emitters, together with the corresponding percentages of the MPC's, in below-outfall creek water are given in Table XVII. The most abundant beta emitter in the creek, in terms of activity, was cobalt-58. The total beta activities in the creek (see Tables XV and XVI) are considerably less than the cobalt-58 concentrations for two reasons. In the decay of cobalt-58, only 15% occurs by positron emission. The portion that decays by electron capture, 85%, contributes little to the measured beta activity. In addition, during 1960 many of the samples containing below-average amounts of beta activity were not analyzed for cobalt, and thus the observed average cobalt-58 (and cobalt-60) activity is greater than the average that would have been obtained if all samples were analyzed for cobalt, or if the samples analyzed were selected at random. During 1961 all Sawmill Creek samples were analyzed, in the form of weekly combined samples, and the average total beta activity

TABLE XVII

BETA EMITTING NUCLIDES IN BELOW-OUTFALL SAWMILL CREEK WATER, 1960-1961

Nuclide	Concentration (pc/liter)				Percent MPC			
	Maximum		Average		Maximum		Average	
	1960	1961	1960	1961	1960	1961	1960	1961
Cobalt-58	2650	735	369	59	2.7	0.7	0.4	0.06
Cobalt-60	171	218	13	8	0.34	0.4	0.03	0.02
Thorium-234	200	42	18	4	1	0.2	0.09	0.02
Cesium-137	83	11.3	7	2.1	0.4	0.06	0.04	0.01
Iodine-131	<2	55	<2	5	<0.1	2.8	<0.1	0.3
Strontium-90	182	9.8	5	1.4	182	9.8	5	1.4
Strontium-89	<1	29.2	<1	3.0	<0.01	0.3	<0.01	0.03
Barium-140	<2	11	<2	2	<0.007	0.04	<0.007	0.007

can be compared directly to 15% of the cobalt-58 activity to obtain the fraction due to this nuclide. The cobalt nuclides are believed to originate primarily in the EBWR reactor at ANL and are produced by neutron irradiation of some of the materials of construction (iron, cobalt, and nickel).

In terms of percent of MPC, the only significant contribution to the beta activity was made by strontium-90, an average of 5% in 1960 and 1.4% in 1961. In 1960 about 60% of the strontium-90 was found in one sample collected on December 9. Excluding this sample, the maximum and average concentrations were 7.0 and 2.1 pc/liter, or 7 and 2.1% of the MPC, respectively. No unusually high strontium-90 concentrations were encountered in 1961, and the average strontium-90 content was correspondingly lower.

The iodine-131 found below the outfall from the middle of February to early April 1961 and above the site in March 1961 was due to the iodine release discussed earlier. Some of this iodine may have entered the creek in ANL waste water. However, some was added directly to the waste water from the atmosphere, since iodine-131 was also found in air, rain, and above-site water during this period.

It should be pointed out that the average values for nuclides found only infrequently in the creek and in concentrations close to the minimum detectable amount may be greater than the true values because of the method used in computing the averages. The nuclides for which this consideration apply, e.g., barium-140 and cesium-137 above the site, can be determined by comparing the maximum and average values in the tables. However, it is probable that the average concentrations of nuclides in samples in which they were not detected is not greater than one-half of the minimum detectable amount, so the average concentration is not underestimated.

As was mentioned previously, the strontium-89 and barium-140 found below the outfall in 1961 were added primarily as fallout. For the other fission products, it is estimated that 20-25% of the iodine-131, cesium-137, and strontium-90 during the year resulted from fallout and that the remainder was added in Argonne waste water. The cobalt nuclides were not detected above the site, so that they were essentially all added in the waste water. During 1960 Argonne added an average of about 33 pcβ/liter to below site water. The remainder was due to fallout (about 1 pc/liter) and natural activity (about 3 pc/liter). During 1961 the Argonne contribution averaged about 20 pc/liter. The decrease resulted principally from a decrease in the amounts of cobalt-58 and thorium-234. The fallout and natural activity contributions in 1961 were about 7 and 3 pc/liter, respectively.

As a result of dilution by ANL waste water, it is possible for the concentrations of some fission products to be lower below the outfall than above. This has been observed in other years and occurred for strontium-89 in December, when the waste water evidently contained less than natural creek water.

The fission product activity during 1960 may be evaluated from the strontium-90 and cesium-137 concentrations in above-site water. The abundances of these nuclides were about the same early in 1960 as in the last quarter of 1959. A slight increase was observed in the spring, probably for the same reasons that the air activities increased, after which a decrease was generally observed. There was little change in the average concentrations of these long-lived nuclides during 1961, in spite of the large increase in short-lived fission products from the 1961 USSR tests, although the fraction of samples giving positive results increased substantially in the last quarter of 1961. Fission products other than those listed in the tables, notably zirconium-95-niobium-95, cerium-141, and cerium-144, were also present at both locations in the creek after September 15, 1961, but since analyses for these nuclides were not done routinely, except qualitatively, the results are not included.

On two occasions water samples were collected at several locations between the outfall and 335 yards downstream to determine if any loss of activity occurred as the water moved downstream. The results are tabulated in Table XVIII. Although the results are not conclusive because the total activities were relatively low, it appears that the losses were small. The decrease in alpha activity in the first 50 yards on October 27, 1961, indicates that any losses occurred early. Unfortunately, the activity at 50 yards (2.4 pcα/liter) was very nearly the same as the natural activity. Studies of the creek bed material in this region of the stream (see Section III-D) show that some activity is absorbed from the water. The rate of removal will certainly vary, even for the same nuclide, with pH, the amount and nature of the creek bed, and other variables. It is planned to repeat this type of sampling and, if losses occur, to determine the identity of the active nuclides in the water and the bed.

2. Des Plaines River

Since Sawmill Creek empties into the Des Plaines River about 500 yards downstream from the waste-water outfall, the river was sampled regularly above and below the mouth of Sawmill Creek to determine if the activity in the creek had any effect on the activity in the Des Plaines River. Prior to May 1960, samples were collected at monthly intervals. Beginning in May, collections were made at weekly intervals when water was available.

TABLE XVIII

TOTAL ACTIVITY IN SAWMILL CREEK AS A FUNCTION OF DISTANCE DOWNSTREAM FROM THE WASTE-WATER OUTFALL

Distance from Outfall (yards)	Nov. 14, 1960		Oct. 27, 1961	
	pcα/liter	pcβ/liter	pcα/liter	pcβ/liter
0	3.6	10.9	6.2	19.4
30	4.6	10.4	-	-
50	-	-	2.4	13.6
80	5.0	11.4	-	-
135	4.9	12.8	-	-
155	-	-	3.1	12.8
220	4.9	13.7		
335	4.8	11.7	3.5	14.3

All samples were analyzed for total activity. Analyses for specific elements and nuclides were performed less frequently, as indicated in the tables.

Alpha and beta activities, and concentrations of uranium and some of the fission products are given in Tables XIX and XX. The activities were very similar at both locations, and no significant or consistent differences could be found.

In addition to the results in the tables, thorium and plutonium analyses were performed monthly; the samples collected in February, March, and April (when iodine-131 released at Argonne was found in air and in Sawmill Creek water) were analyzed for iodine-131. None of the samples analyzed contained detectable amounts of these activities.

Gamma-ray spectra were taken on all samples, and cobalt separations were performed occasionally. Cobalt activities were not detected, and the gamma-ray spectra were very similar at both locations.

All of the results indicate that the activity in Sawmill Creek had no effect on the radioactivity in the Des Plaines River. This is reasonable in view of the large dilution of Sawmill Creek water by the much greater volume of water in the river.

The alpha activities and uranium concentrations throughout both years were similar to those found earlier in the river and are considered normal. The beta activities during 1960 and the first eight months

of 1961 varied between about 5 and 15 pc/liter and averaged approximately 8 pc/liter. Beta activities of this magnitude have been found in the past when fallout activity was small compared with the natural activity, and are considered to be essentially normal for the river.

TABLE XIX

RADIOACTIVITY IN DES PLAINES RIVER WATER, 1960
(pc/liter)

Date Collected[1]	Location[2]	Alpha Activity	Beta Activity	Uranium	Cesium-137	Strontium-90
February 28	B	2.0	5.9	2.1	-	0.8
March 30	A	2.4	7.5	1.0	-	-
	B	2.3	7.5	0.94	-	-
April 27	A	1.8	3.7	1.3	< 0.5	-
	B	1.9	6.4	1.3	< 0.5	-
May	A	2.5	8.3	1.3	-	1.1
	B	2.5	8.5	1.4	< 0.5	1.1
June	A	2.7	7.3	1.5	0.7	-
	B	2.6	8.9	1.6	< 0.5	1.3
July	A	2.4	7.4	1.5	< 0.5	< 0.5
	B	2.3	7.8	1.8	< 0.5	< 0.5
August	A	2.8	7.6	1.6	-	-
	B	2.1	8.2	1.6	-	-
September	A	1.5	9.4	1.2	< 0.5	< 0.5
	B	1.5	10.1	1.3	< 0.5	1.0
October	A	1.2	11.7	1.6	1.1	-
	B	1.7	10.6	1.0	-	< 0.5
November	A	3.1	11.6	1.5	-	0.9
	B	2.6	11.2	1.5	-	0.8
December	A	3.2	9.3	1.5	-	-
	B	2.6	7.9	1.6	-	-
Average	A	2.4	8.4	1.4	0.5	0.6
	B	2.2	8.5	1.5	0.3	0.9

[1] From May through December samples were collected weekly. All samples were analyzed for total alpha, beta, and uranium activities. The averages are given in the table. The maximum values from May through December were:

	Alpha Activity	Beta Activity	Uranium
Location A	4.2	15.9	2.3
Location B	3.5	13.5	2.2

The other analyses were performed on one sample each month and are listed individually in the table.

[2] Location A is approximately 5 miles above the mouth of Sawmill Creek. Location B is approximately 2.3 miles below the mouth of Sawmill Creek.

TABLE XX

NONVOLATILE RADIOACTIVITY IN DES PLAINES RIVER WATER, 1961

Month	Location[1]	Alpha Activity (pc/liter)			Beta Activity (pc/liter)			Uranium (pc/liter)			Strontium-90 (pc/liter)			Strontium-89 (pc/liter)			Barium-140 (pc/liter)			Cesium-137 (pc/liter)		
		No. of Samples	Max	Av	No. of Samples	Max	Av	No. of Samples	Max	Av	No. of Samples	Max	Av[2]	No. of Samples	Max	Av[2]	No. of Samples	Max	Av[2]	No. of Samples	Max	Av[2]
January	A	1	2.2	-	1	10.5	-	1	1.7	-	0	-	-	0	-	-	0	-	-	1	<0.5	-
	B	1	2.6	-	1	10.4	-	1	2.2	-	0	-	-	0	-	-	0	-	-	1	<0.5	-
February	A	2	1.9	1.6	2	13.8	12.4	1	1.0	-	2	<0.5	-	2	<2	-	2	<2	-	1	<0.5	-
	B	2	2.2	1.7	2	14.0	12.3	2	1.5	1.5	1	0.6	-	1	<2	-	1	<2	-	1	<0.5	-
March	A	5	4.1	2.5	5	11.8	8.0	2	1.3	1.2	1	0.5	-	1	<2	-	1	<2	-	1	<0.5	-
	B	5	2.8	2.3	5	11.0	8.2	5	1.6	1.3	1	0.8	-	1	<2	-	1	<2	-	1	<0.5	-
April	A	4	2.5	2.2	4	10.6	8.4	3	2.1	1.9	1	0.8	-	1	<2	-	1	<2	-	1	<0.5	
	B	4	3.2	2.4	4	9.8	7.4	4	2.3	2.0	1	0.7	-	1	<2	-	1	<2	-	1	<0.5	-
May	A	5	2.8	2.3	5	9.3	7.7	2	1.9	1.7	1	<0.5	-	1	<2	-	1	<2	-	1	<0.5	-
	B	5	3.3	1.9	5	11.3	9.2	5	1.7	1.5	1	<0.5	-	1	<2	-	1	<2	-	1	<0.5	-
June	A	3	5.0	2.4	3	16.0	11.5	1	1.2	-	1	0.9	-	1	<2	-	1	<2	-	1	0.5	-
	B	4	3.2	2.5	4	13.3	10.3	1	2.2	1.5	1	0.8	-	1	<2	-	1	<2	-	1	0.6	-
July	A	4	2.4	1.9	4	14.2	11.6	2	1.4	1.1	1	0.9	-	1	<2	-	1	<2	-	1	<0.5	-
	B	4	2.0	1.4	4	12.2	10.9	3	1.2	1.1	1	1.3	-	1	<2	-	1	<2	-	1	<0.5	-
August	A	5	5.1	2.6	5	15.8	12.3	1	1.0	-	1	1.0	-	1	<2	-	1	<2	-	1	<0.5	-
	B	5	2.7	2.0	5	11.7	10.4	5	1.2	0.9	1	1.4	-	1	<2	-	1	<2	-	1	0.6	-
September	A	4	3.8	2.2	4	22.5	14.6	2	1.1	0.9	1	0.6	-	1	5.5	-	1	4.3	-	1	0.9	-
	B	4	3.7	2.6	4	21.3	16.1	4	2.6	1.2	1	0.5	-	1	4.8	-	1	4.2	-	1	<0.5	-
October	A	4	4.4	3.3	4	22.5	18.9	2	2.2	1.9	1	0.5	-	1	2.2	-	1	3.9	-	1	1.0	-
	B	4	5.5	3.5	4	26.3	21.2	4	2.3	1.8	1	0.6	-	1	4.7	-	1	4.3	-	1	2.5	-
November	A	5	3.8	2.5	4	48.1	37.0	2	1.7	1.6	1	0.8	-	1	14.2	-	1	11.1	-	1	0.9	-
	B	5	3.1	2.3	4	54.0	40.5	5	1.7	1.5	1	0.5	-	1	13.5	-	1	6.3	-	1	0.5	-
December	A	3	5.1	4.2	3	66.0	57.1	2	2.0	1.9	1	0.8	-	1	10.2	-	1	3.3	-	1	0.5	-
	B	4	3.8	3.3	4	56.1	47.8	4	2.1	1.9	1	0.8	-	1	13.6	-	1	3.7	-	1	1.7	-
Annual Summary	A	45	5.1	2.5	45	66.0	17.5	21	2.2	1.5	12	1.0	0.7	12	14.2	3.0	12	11.1	2.2	12	1.0	0.5
	B	47	5.5	2.4	47	56.1	17.1	43	2.3	1.5	11	1.4	0.8	11	13.6	3.6	11	6.3	2.0	12	2.5	0.6

[1]Location A is approximately 5 miles above the mouth of Sawmill Creek.
Location B is approximately 2.3 miles below the mouth of Sawmill Creek.

[2]The averages were calculated assuming a concentration of one-half of the minimum detectable concentration for those samples in which a given nuclide was not detected.

The effect of the presence of recently produced fission product fallout on the total beta, barium-140, and strontium-89 concentrations after September 1961 can be seen from Table XX. Short-lived fission products could not be detected prior to September 1961. The average fission product concentrations were calculated as indicated previously for Sawmill Creek, using one-half of the minimum detectable amount as the concentration in samples containing less than this minimum. This results in an overestimated average for the short-lived fission products, barium-140 and strontium-89, since their concentrations were probably much less than the one-half value prior to September. If the concentrations of these nuclides are assumed to have been zero prior to September, the annual averages are about 10% lower than those in the table. During the four months in which these nuclides were detected, the concentrations were 8.0 and 5.7 pc/liter for strontium-89 and barium-140, respectively, above the creek, and 9.2 and 4.7 pc/liter, respectively, below the creek.

3. Other Waters

The total activities in other streams and ponds on the ANL site are given in Table XXI. The above-average activity in the storage lagoon for contaminated waste water resulted from the activity in the waste water stored occasionally in the lagoon and from activity

TABLE XXI

NONVOLATILE RADIOACTIVITY IN PONDS AND STREAMS ON ANL SITE, 1960-61

Date Collected	No. of Samples	Alpha Activity (pc/liter)		Beta Activity (pc/liter)	
		Max	Av	Max	Av
April 29, 1960	3	4.4[a]	3.1	105[a]	40
	2[b]	2.8	2.5	7.7	7.5
July 29, 1960	3	1.9	1.2	8.5	6.9
September 30, 1960	2	5.2	3.0	7.7	7.5
November 21, 1960	4	5.8	2.9	21.4[a]	10.8
	3[b]	5.8	2.5	9.3	7.3
December 7, 1960	1	2.1	-	5.7	-
August 29, 1961	3	3.9[a]	2.4	61.5[a]	38.8
	2[b]	2.3	1.7	8.4	8.1
September 22, 1961	4	1.9	1.5	16.3	13.5
Summary	20	5.8	2.3	105	19
	17[b]	5.8	1.9	16.3	8.7

[a]Collected from lagoon used for the storage of contaminated waste water.

[b]Excluding storage lagoon for contaminated waste water.

leached from the bed of the lagoon. The bed contains activity absorbed from waste water stored in previous years, so that lagoon water becomes contaminated when only rain is present in the lagoon. The same nuclides found in Sawmill Creek due to waste water were found in the lagoon (cobalt-58, cobalt-60, strontium-90, cesium-137, uranium, and plutonium).

Similar results were obtained in other years. The total activities in the natural ponds were similar to those found earlier at the same locations. Samples from one lagoon, at location 11G in Figure 13, contained up to about 6 pc/liter of alpha activity. This activity was due principally to uranium. The average alpha activity in surface water is approximately 2 pcα/liter. The beta activities in the natural ponds prior to September 1961, 5-10 pc/liter, were uniformly low and close to natural levels. The increase in the samples collected on September 22, 1961, was due to fallout from the USSR tests. The beta activity in these samples decayed as expected.

The total activities in lakes and streams within 25 miles of the ANL site are listed individually in Tables XXII and XXIII and summarized in Table XXIV. The average and range (0.3 to 4.7 pc/liter) of alpha activities were normal through both years. Except for one sample collected from Lake Calumet in July 1960, the beta activities were relatively low prior to September 1961 and were similar to those found in the Des Plaines River. The average beta activity, about 8 pc/liter, was smaller than for any year since sample collection was begun in 1952. Previous annual averages ranged from 15 pc/liter in 1954 to 43 pc/liter in 1953. These fluctuations were due to varying amounts of fission product activity,

TABLE XXII

NONVOLATILE RADIOACTIVITY IN SURFACE WATER NEAR ANL, 1960

Location	April		May		July		September		November	
	pcα/g	pcβ/g	pcα/g	pcβ/g	pcα/g	pcβ/g	pcα/g	pcβ/g	pcα/g	pcβ/g
Des Plaines River, Brookfield	1.5	6.1	-	-	-	-	2.0	7.0	3.8	6.0
Des Plaines River, Willow Springs	1.8	3.7	2.5	9.7	1.9	7.8	1.5	7.8	3.4	10.0
Des Plaines River, Lemont	1.9	6.4	2.4	9.4	1.8	8.2	1.6	10.4	2.7	7.1
Des Plaines River, Romeoville	-	-	-	-	1.9	7.7	1.3	8.1	2.6	8.3
Illinois River, Morris	1.8	4.9	1.4	4.7	-	-	-	-	2.3	5.9
Confluence - Illinois, Kankakee, and Des Plaines Rivers	-	-	2.7	8.3	4.6	14.2	1.6	6.7	1.6	6.5
Du Page River, Naperville	2.5	4.8	-	-	-	-	1.7	12.6	3.1	14.9
Du Page River, Channahon	-	-	-	-	-	-	1.5	7.6	2.3	6.8
Flag Creek, German Church and Wolf Roads	-	-	-	-	1.0	10.0	-	-	-	-
Salt Creek, Wolf Road, Western Springs	-	-	-	-	-	-	-	-	4.7	12.8
McGinnis Slough, U. S. Route 45 and Ill. Route 7	-	-	-	-	-	-	-	-	1.9	14.8
Saganashkee Slough, 104th Ave. and Sag Canal	0.7	8.6	-	-	-	-	-	-	-	-
Sanitary and Ship Canal, Lemont	-	-	-	-	0.7	5.8	-	-	0.4	4.5
Fox River, Aurora	-	-	-	-	-	-	-	-	1.0	3.0
Lake Calumet, 111th St., Chicago	-	-	-	-	0.9	76.1	-	-	1.6	5.2
Lake Michigan, 111th St., Chicago	-	-	-	-	0.3	1.3	-	-	0.4	1.4
Cal-Sag Canal, 109th and Willow Springs Road	-	-	-	-	-	-	-	-	1.1	5.0
Average	1.7	5.8	2.3	8.0	1.6	16.4	1.6	8.6	2.2	7.5

TABLE XXIII

NONVOLATILE RADIOACTIVITY IN LAKES AND STREAMS NEAR ANL, 1961

Location	April		May		August		September		October	
	pcα/g	pcβ/g	pcα/g	pcβ/g	pcα/g	pcβ/g	pcα/g	pcβ/g	pcα/g	pcβ/g
Des Plaines River, Brookfield	2.0	7.2	-	-	1.2	11.9	-	-	-	-
Des Plaines River, Willow Springs	2.2	8.4	2.3	7.7	2.6	12.3	2.2	14.6	3.3	18.9
Des Plaines River, Lemont	2.4	7.4	1.9	9.2	2.0	10.4	2.6	16.1	3.5	21.2
Des Plaines River, Romeoville	1.3	6.0	-	-	1.1	8.9	2.5	11.3	-	-
Illinois River, Morris	-	-	1.3	3.8	-	-	-	-	3.0	19.8
Confluence - Illinois, Kankakee, and Des Plaines Rivers	2.4	8.2	1.0	5.4	1.4	5.8	2.0	8.5	-	-
Du Page River, Naperville	2.2	6.9	-	-	-	-	-	-	-	-
Du Page River, Channahon	-	-	-	-	1.2	8.1	-	-	-	-
Flag Creek, German Church and Wolf Roads	-	-	-	-	1.0	20.2	-	-	-	-
Salt Creek, Wolf Road, Western Springs	2.3	7.6	-	-	-	-	-	-	-	-
McGinnis Slough, U. S. Route45 & Ill. Route7	1.5	14.1	-	-	-	-	-	-	-	-
Cal-Sag Canal	-	-	-	-	-	-	1.3	10.7	-	-
Saganashkee Slough, 104th Ave. and Sag Canal	-	-	-	-	-	-	0.3	6.8	-	-
Sanitary and Ship Canal, Lemont	-	-	-	-	0.3	4.5	-	-	-	-
Fox River, Aurora	0.3	4.1	-	-	-	-	-	-	-	-
Sauk Lake, Park Forest	-	-	-	-	-	-	0.3	5.8	-	-
Lake Michigan, 111th St., Chicago	-	-	-	-	-	-	0.2	1.9	-	-
Long Run Creek	-	-	-	-	-	-	1.3	5.4	-	-
Average	1.8	7.8	1.6	6.5	1.4	10.3	1.4	9.0	3.3	20.0

TABLE XXIV

NONVOLATILE RADIOACTIVITY IN SURFACE WATER NEAR ANL, 1960-61

Month	Distance from ANL (miles)	No. of Samples	Alpha Activity (pc/liter)		Beta Activity (pc/liter)	
			Max	Av	Max	Av
April, 1960	10	4	2.5	1.7	6.4	5.9
	25	2	1.8	1.7	6.1	5.5
May, 1960	10	2	2.5	2.4	9.7	9.6
	25	2	2.7	2.1	8.3	6.5
July, 1960	10	5	1.9	1.5	10.0	7.9
	25	3	4.6	1.9	76.1	30.2
September, 1960	10	4	1.7	1.5	12.6	9.7
	25	3	2.0	1.7	7.6	7.1
November, 1960	10	8	4.7	2.5	14.9	9.7
	25	7	3.8	1.9	6.8	5.0
1960 Summary	10	23	4.7	2.0	14.9	8.6
	25	17	4.6	1.9	76.1	10.1
	10 and 25	40	4.7	2.0	76.1	9.0
April, 1961	10	4	2.3	2.0	14.1	9.0
	25	3	2.4	1.3	8.2	6.1
May, 1961	25	2	1.3	1.2	5.4	4.6
August, 1961	10	3	1.2	0.8	20.2	12.3
	25	3	1.4	1.2	8.1	7.6
September, 1961	10	3	1.3	1.0	10.7	7.6
	25	4	2.5	1.3	11.3	6.9
October, 1961	25	1	3.1	-	19.8	-
1961 Summary	10	10	2.3	1.5	20.2	9.6
	25	13	3.1	1.4	19.8	7.5

and the exact average obtained depends on the times of sample collection and nuclear testing, since these samples are collected only at several month intervals. Samples collected in September and October 1961 showed the effect of fallout from the 1961 tests. The beta activities in some of the samples increased by up to a factor of two, and the beta activity decayed by about a factor of two in four months. The amount of fresh fallout varied considerably between samples. At some locations the presence of short-lived fission products could not be detected from the total beta activity and the rate of beta decay.

The Lake Calumet sample taken in July 1960 contained 76 pc/liter of beta activity, about 10 times the normal value for this location. This sample showed no beta decay over a period of eight months and did not contain strontium-90 or any detectable amounts of gamma-emitting nuclides. The source and identity of the activity in this sample is not known.

Additional analyses were performed on about one-third of the Illinois River samples with the following results. Strontium-90 and cesium-137 concentrations ranged from 0.5 to 0.7 pc/liter, uranium from 0.8 to 2.2 pc/liter, thorium from less than 0.05 to 0.16 pc/liter, and the plutonium concentrations were all less than 0.05 pc/liter. The Illinois River samples are of interest because the Des Plaines River (containing Sawmill Creek water) meets the Kankakee River to form the Illinois, and the Dresden Nuclear Power Plant is located at this junction. No activity other than fallout and natural activity was found in any of these samples.

The total activities in the reference site samples are given in Table XXV. The alpha activities were uniformly low and normal. The variations in beta activity were similar to those found in water collected near ANL. The beta activities were low in May and November 1960 and in May 1961. The effect of fallout on the October 1961 samples is evident from the total beta activities in all samples, including those from Lake Michigan. The beta activity in all of these samples decayed appreciably in a few months, confirming the presence of short-lived fission products.

TABLE XXV

NONVOLATILE RADIOACTIVITY IN SURFACE WATER FROM REFERENCE SITES, 1960-61

Location	May 25-26, 1960		November 16-17, 1960		May 25-26, 1961		October 24-26, 1961	
	pcα/liter	pcβ/liter	pcα/liter	pcβ/liter	pcα/liter	pcβ/liter	pcα/liter	pcβ/liter
Lake Delavan, Wisconsin	0.5	5.0	1.8	4.8	0.7	3.2	0.6	19.3
Fox River, Oak Point State Park, Wisconsin	0.6	4.9	2.1	6.6	0.3	2.0	0.9	13.5
Lake Michigan, St. Joseph, Michigan								
near shore	0.2	1.6	0.3	0.8	0.2	1.2	0.1	3.2
1500 ft from shore	-	-	-	-	0.09	1.2	0.1	2.5
Magician Lake, Michigan	0.1	4.2	0.4	3.5	0.1	3.1	0.1	9.7
Illinois River, Starved Rock State Park, Illinois	1.4	4.2	1.7	7.4	0.9	4.8	3.1	16.4
Shafer Lake, Monticello, Indiana	1.2	2.8	1.6	2.3	1.2	1.9	0.9	9.2
Kankakee River, Kankakee River State Park, Indiana	0.7	2.0	-	-	0.9	2.3	-	-
Average	0.7	3.5	1.3	4.2	0.5	2.5	0.8	10.5

The activity due to fallout has been appreciably higher in other years. For example, in 1958 and 1959 the beta activities in Lake Michigan varied from 7 to 21 pc/liter and in Lake Delavan from 7 to 50 pc/liter as a result of fallout.

D. Bottom Silt

The activity in the beds of lakes and streams is of interest for several reasons. Where the conditions are appropriate for removing and concentrating activity from water, plant and animal life may be exposed to higher radiation doses than expected from water activities alone. The bed, by concentrating activity, may show low-level stream contamination when water analyses do not. The bed can also retain activity for a considerable length of time, and indicate water contamination that was undetected in the past.

The total activities in samples of bottom silt collected monthly from Sawmill Creek and the Des Plaines River are given in Tables XXVI and XXVII. Although total activities in bottom silt vary considerably between locations and between samples collected at different times at the same location, alpha activities in excess of 35 pc/g and beta activities in excess of 90 pc/g are abnormally high. In borderline cases, specific analyses are

TABLE XXVI

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT FROM SAWMILL CREEK AND DES PLAINES RIVER, 1960

Date Collected	Location*	Sawmill Creek		Des Plaines River	
		pcα/g	pcβ/g	pcα/g	pcβ/g
January 21	B	51	113	-	-
March 30	A	23	65	21	72
	B	26	47	22	87
April 6	A	22	73	22	64
	B	30	106	37	65
May 4	A	20	53	21	67
	B	26	74	24	86
July 6	A	22	60	21	64
	B	20	67	23	87
August 3	A	26	66	24	89
	B	25	52	25	68
September 7	A	23	65	20	64
	B	50	173	25	75
October 7	A	27	64	21	61
	B	17	34	25	65
November 2	A	26	59	20	59
	B	16	33	26	61
December 7	A	27	60	18	37
	B	17	35	26	50
Average	A	24	63	21	64
	B	28	86	26	72

*Sawmill Creek Locations were:
A - Above ANL Site (location 13L in Figure 13).
B - Below the waste water outfall (location 7M in Figure 13).

Des Plaines River Locations were:
A - Willow Springs, approximately 5 miles above the mouth of Sawmill Creek.
B - Lemont, approximately 2.3 miles below the mouth of Sawmill Creek.

TABLE XXVII

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT FROM SAWMILL CREEK AND DES PLAINES RIVER, 1961

Date Collected	Location*	Sawmill Creek		Des Plaines River	
		pcα/g	pcβ/g	pcα/g	pcβ/g
February 22	A	24	51	23	47
	B	24	41	24	56
March 7	A	24	57	17	35
	B	26	54	26	62
April 5	A	16	37	20	48
	B	25	41	21	65
June 7	A	23	51	23	46
	B	55	106	28	58
July 5	A	26	60	27	45
	B	60	104	28	56
August 2	A	29	53	19	32
	B	51	175	30	61
September 6	A	30	55	19	35
	B	23	67	25	46
October 4	A	29	50	34	112
	B	19	40	28	68
November 1	A	29	49	22	53
	B	29	69	28	77
December 6	A	24	59	25	69
	B	29	78	28	216
Average	A	25	52	23	52
	B	34	78	27	77

*Sawmill Creek Locations were:
A - Above ANL Site (location 13L in Figure 13).
B - Below the waste water outfall (location 7M in Figure 13).

Des Plaines River Locations were:
A - Willow Springs, approximately 5 miles above the mouth of Sawmill Creek.
B - Lemont, approximately 2.3 miles below the mouth of Sawmill Creek.

required to determine the absence or presence of non-natural activities. About one-fourth of the monthly samples collected from Sawmill Creek below the outfall contained above-normal alpha and beta activities due to radioactive contamination in Argonne waste water.

The monthly samples were collected about 10 yards from the outfall. The creek bed was also sampled in the first 335 yards below the outfall to determine the extent of the contamination. The results are given in Table XXVIII. These activities varied greatly with time and location, indicating that activity in the creek water can be carried considerable distances before it is adsorbed and that the nature of the bed material differs from place to place. The samples collected on October 27, 1961, gave uniformly normal results, while the survey made in 1960 showed several peaks. A similar variation with distance was found in a survey made in 1958.

TABLE XXVIII

NONVOLATILE RADIOACTIVITY IN BED OF SAWMILL CREEK

Distance Downstream from Waste Water Outfall (yards)	Alpha Activity (pc/g)		Beta Activity (pc/g)	
	Nov. 14, 1960	Oct. 27, 1961	Nov. 14, 1960	Oct. 27, 1961
0	19	20	46	51
10	209	28	502	45
20	82	30	197	41
30	63	16	123	34
40	17	12	46	24
50	242	23	479	44
60	247	27	439	41
100	20	10	42	21
135	19	23	45	61
155	197	14	250	28
220	20	16	38	32
260	78	9	109	23
295	38	14	66	28
310	29	25	47	46
335	26	21	42	44

The same nuclides added to the creek in Argonne waste water contributed to the increased activity in below-outfall bottom silt samples, as shown in Table XXIV. Normal concentrations of uranium and thorium in bottom silt from the Chicago area are 1-3 pc/g based on previous analyses. The plutonium, strontium-90, and cesium-137 contents of bottom silt colected in 1959 from the Chicago area were < 0.1, 0.2-0.5, and 2-4 pc/g, respectively. Since the amount of fallout decreased sharply in 1960, these plutonium, strontium-90, and cesium-137 concentrations may be considered as present in the 1960 samples due to fallout. Thus, concentrations in excess of those given above were derived from Argonne waste water, and, as shown in Table XXIX, samples containing abnormally high alpha and beta activities also contained high concentrations of these nuclides.

TABLE XXIX

RADIOACTIVITY (pc/g) IN SELECTED BOTTOM SILT SAMPLES, 1960

Location	Date	Alpha	Beta	Uranium	Thorium	Plutonium	Strontium-90	Cesium-137
Sawmill Creek, 10 yards below outfall	September 7	50	173	11	-	-	-	-
Sawmill Creek, 10 yards below outfall	November 14	209	502	49	11	20	2.7	5.4
Sawmill Creek, 80 yards below outfall	November 14	247	493	40	-	-	3.9	2.7
Sawmill Creek, 155 yards below outfall	November 14	197	250	26	-	12	2.2	1.7
Sawmill Creek, 295 yards below outfall	November 14	38	66	3.6	0.85	2.7	<0.2	1.6
Des Plaines River, Lemont (below Sawmill Creek)	April 6	37	65	2.3	-	-	-	-
Illinois River, Morris	April 28	19	35	1.1	-	-	-	-
Lake Calumet, Chicago	November 17	32	65	3.1	-	-	-	-
Storage Lagoon, ANL	November 21	242	775	25	-	-	-	-

Bottom silt from Sawmill Creek above the site contained normal alpha and beta activities. Previous annual averages, 22-29 pcα/g and 60-93 pcβ/g, were similar to those in 1960 and 1961. Increases in the fall of 1961 due to fallout were small. Total alpha and beta activities in the Des Plaines River samples were in the normal range except during the last three months of 1961, when increases in beta activity due to recently produced fallout were detected. Differences between the two locations occurred in both directions in a random manner and could not be attributed to activity entering the river from Sawmill Creek. Thus, the sample collected on April 6, 1960, below Sawmill Creek contained 37 pcα/g, slightly higher than the usual alpha activity. However, the uranium content (2.3 pc/g) was in the normal range, plutonium and thorium were not detected, and the ratio of alpha activity to uranium concentration was normal. In addition, the sample contained normal amounts of beta activity, and since Sawmill Creek samples containing alpha activity from Argonne waste water have also contained elevated beta activities, Des Plaines River samples should behave similarly. The radioactivity in the April 6 Des Plaines River sample must, therefore, be considered normal. The high beta activities in October and December samples were due to above-normal amounts of fallout at the particular locations sampled. These samples contained the same fission products found in air during this period.

Table XXX gives the total activities found in other ponds and streams on the ANL site. Abnormally high activities were consistently found in samples from the storage lagoon for contaminated waste water.

TABLE XXX

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT FROM OTHER PONDS AND STREAMS ON ANL SITE, 1960-61

Date Collected	No. of Samples	Alpha Activity (pc/g)		Beta Activity (pc/g)	
		Max	Av	Max	Av
April 29, 1960	3	325[a]	124	392[a]	179
	2[b]	26	24	75	73
July 28, 1960	3	26	25	54	48
September 30, 1960	2	28	25	63	58
November 21, 1960	4	242[a]	77	775[a]	228
	3[b]	23	22	48	46
August 29, 1961	3	30	29	54	51
September 22, 1961	4	46	31	58	47
Summary	19	325[a]	53	775	108
	17	46	26	75	52

[a]Sample collected from storage lagoon for contaminated waste water.

[b]Excluding samples collected from storage lagoon.

One sample, collected from a pond on the southern edge of the ANL site on September 22, 1961, contained an above-average alpha activity, 46 pc/g. Some of the previous samples from this pond gave similar results, and these samples contained above average concentrations of thorium-232. The thorium probably occurs naturally and cannot be correlated with the thorium usage at Argonne. Fallout from the USSR tests in 1961 did not increase the beta activity in the September 1961 samples significantly.

The total activities in bottom silt within 25 miles of the Laboratory and at the reference sites are given in Tables XXXI to XXXIV. The alpha and beta activities were normal and in the range found previously at each location. The beta activities in the samples collected in the fall of 1961 were not significantly affected by fallout from the 1961 nuclear tests, although this fallout was found in water samples from some of these locations.

TABLE XXXI

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT NEAR ANL, 1960

Location	April		May		July		September		November	
	pcα/g	pcβ/g	pcα/g	pcβ/g	pcα/g	pcβ/g	pcα/g	pcβ/g	pcα/g	pcβ/g
Des Plaines River, Brookfield	20	75	-	-	-	-	18	40	26	51
Des Plaines River, Willow Springs	22	64	21	67	21	64	20	64	20	59
Des Plaines River, Lemont	37	65	24	86	23	87	25	75	26	61
Des Plaines River, Romeoville	17	50	-	-	19	60	22	58	22	54
Illinois River, Morris	19	35	5.5	27	-	-	-	-	4.1	17
Du Page River, Naperville	-	-	-	-	-	-	-	-	50	70
Du Page River, Channahon	-	-	-	-	-	-	20	36	25	43
Flag Creek, German Church and Wolf Roads	-	-	-	-	30	58	-	-	-	-
Salt Creek, Wolf Road, Western Springs	-	-	-	-	-	-	-	-	32	60
McGinnis Slough, U. S. Route 45 and Illinois Route 7	-	-	-	-	-	-	-	-	27	44
Saganashkee Slough, 104th Ave. and Sag Canal	20	49	-	-	-	-	-	-	-	-
Fox River, Aurora	-	-	-	-	-	-	-	-	7.2	20
Lake Calumet, 111th St., Chicago	-	-	-	-	25	31	-	-	32	65
Average	23	56	17	60	24	60	21	55	25	49

TABLE XXXII

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT NEAR ANL, 1961

Location	March		April		August		September	
	pcα/g	pcβ/g	pcα/g	pcβ/g	pcα/g	pcβ/g	pcα/g	pcβ/g
Des Plaines River, Brookfield	-	-	25	53	18	42	22	48
Des Plaines River, Willow Springs	17	35	20	48	19	32	-	-
Des Plaines River, Lemont	26	62	21	65	30	61	-	-
Des Plaines River, Romeoville	-	-	28	51	26	45	-	-
Illinois River, Morris	-	-	11	22	-	-	3.7	20
Du Page River, West Chicago	28	32	-	-	-	-	-	-
Du Page River, Naperville	-	-	40	51	34	45	-	-
Flag Creek, German Church and Wolf Roads	-	-	-	-	32	44	-	-
Salt Creek, Wolf Road, Western Springs	-	-	22	46	-	-	-	-
McGinnis Slough, U. S. Route 45 and Ill. Route 7	-	-	29	53	-	-	-	-
Saganashkee Slough, 104th Ave. and Sag Canal	-	-	-	-	-	-	40	42
Fox River, Aurora	-	-	18	29	-	-	-	-
Long Run Creek, 135th Ave. and Ill. Route 4A	-	-	-	-	-	-	21	50
Average	24	43	24	46	27	45	22	40

TABLE XXXIII

NONVOLATILE RADIOACTIVITY IN BOTTOM SILT NEAR ANL, 1960-61

Month	Distance from ANL (miles)	No. of Samples	Alpha Activity (pc/g)		Beta Activity (pc/g)	
			Max	Av	Max	Av
April, 1960	10	3	37	21	65	53
	25	3	20	20	75	53
May, 1960	10	2	24	23	86	77
	25	1	55	-	27	-
July, 1960	10	3	30	25	87	70
	25	2	25	22	60	46
September, 1960	10	3	25	21	75	60
	25	2	22	21	58	47
November, 1960	10	6	32	26	61	55
	25	5	50	24	70	43
1960	10	17	37	23	87	61
Summary	25	14	50	21	70	46
	10 and 25	31	50	22	87	55
March, 1961	10	2	26	22	62	48
	25	2	36	28	37	32
April, 1961	10	5	29	22	65	53
	25	4	40	27	53	39
August, 1961	10	4	30	27	34	26
	25	2	61	45	45	44
September, 1961	10	2	40	31	48	45
	25	2	22	13	48	34
1961	10	13	40	25	65	43
Summary	25	10	61	28	53	38
	10 and 25	23	61	26	65	41

TABLE XXXIV

RADIOACTIVITY IN BOTTOM SILT FROM REFERENCE SITES, 1960-61

Location	May 25-26, 1960		November 16-18, 1960		May 24-25, 1961		October 24-25, 1961	
	pcα/g	pcβ/g	pcα/g	pcβ/g	pcα/g	pcβ/g	pcα/g	pcβ/g
Lake Delavan, Wisconsin	2.5	25	3.0	15	14	26	2.1	19
Fox River, Oak Point State Park, Wisconsin	6.6	32	19	37	19	40	4.6	26
Magician Lake, Michigan	5.5	26	4.0	18	6.0	29	4.1	25
Lake Michigan, St. Joseph, Michigan	-	-	1.7	19	-	-	-	-
Starved Rock State Park, Illinois	1.0	2.9	2.6	1.7	5.7	4	1.2	4.6
Shafer Lake, Indiana	8.0	36	5.5	21	4.0	18	4.4	27
Kankakee River, Kankakee River State Park, Indiana	15	34	-	-	-	-	-	-
Average	6.7	26	6.0	19	10	23	3.5	20

The above-average activities in the bed of the Du Page River at Naperville have been observed previously and are due to thorium-232. The uranium concentrations in this bed have always been normal. The wide variation in both alpha and beta activities shown in the tables illustrate the large natural range of activities in bottom silt. In general, samples composed largely of sand or organic matter contain less activity than samples composed largely of clay.

E. Surface Soil

The total activities in soil on the ANL site during 1960 and 1961 are given in Table XXXV. Normal activities range from approximately 10 to 30 pc/g for alpha activity and from 20 to 80 pc/g for beta activity. The lower values are generally encountered in sandy soils while soils containing clay and loam are more active. As indicated in the table, abnormally high activities were present in samples collected near a uranium-storage shed and near the storage lagoon for contaminated waste water. These activities result from the use made of these areas.

TABLE XXXV

NONVOLATILE RADIOACTIVITY IN SURFACE SOIL ON ANL SITE, 1960-61

Date Collected	No. of Samples	Alpha Activity (pc/g)		Beta Activity (pc/g)	
		Max	Av	Max	Av
April 29, 1960	3	24	20	69	57
July 26, 1960	12[a]	6100	606	4340	483
July 28, 1960	1	25	-	61	-
August 16, 1960	8[b]	26	22	81	54
August 19, 1960	1[c]	31	-	55	-
October 3, 1960	12[c]	47	26	82	55
November 21, 1960	6	28	22	55	47
	1[c]	74	-	248	-
1960 Summary	44	6100	187	4340	178
	31[d]	47	24	82	53
March 10, 1961	4	38	27	70	57
March 29, 1961	5	31	27	70	62
March 30, 1961	11	27	23	62	53
May 11, 1961	4	32	23	54	45
June 23, 1961	5[a]	40	26	91	63
August 29, 1961	9	41[c]	26	75	56
September 22, 1961	2	25	23	62	56
1961 Summary	40	41	25	86	56

[a]Collected near uranium-storage shed.
[b]Collected near EBWR and CP-5 Reactor Buildings.
[c]Collected near storage lagoon for contaminated waste water.
[d]Excluding uranium-storage area and November 21 sample from lagoon area.

Additional analyses confirmed the presence of uranium in soil collected near the storage shed. Surface soil in the Chicago area normally contains about 2 pc of uranium per gram, while the uranium content of the soil near the storage shed ranged from normal amounts to about 6000 pc/g.

Abnormal uranium concentrations were detected up to 75 ft from the shed, an increase over the 40-ft radius of contamination found earlier.

The abnormally high activities in soil collected near the storage lagoon were due to the same nuclides added to Sawmill Creek in Argonne waste water. The contamination was confined to an area that carries overflow lagoon water to Sawmill Creek. At other locations near the lagoon the soil activities were normal.

Iodine-131 was detected in surface soil collected near the building from which the bulk of the iodine was released during February and March 1961. This release, and the resulting airborne iodine-131 concentrations, are discussed in Section II-A. The radioiodine concentrations in soil ranged from less than 0.3 to 104 pc/g at the time of collection, March 29-30. The results are tabulated in Section F in order to compare them with the concentrations in grass collected at the same time. The presence of iodine-131 is not apparent from the results given in Table XXXV since these activities were determined after the samples were dried at 110°C. Drying for several hours at this temperature removed essentially all of the iodine-131. The iodine-131 concentrations were determined on separate, undried portions of the samples.

Other samples collected on the ANL site during 1960 and 1961 contained normal amounts of activity. Recent fallout activity was not apparent in the samples collected on September 22, 1961.

The total activities in soil collected within 25 miles of the ANL site are given in Table XXXVI. The average alpha activity, about 20 pc/g, was normal, and the average beta activity, about 50 pc/g, was very similar to that found in other years when fallout activity was low.

Several samples were analyzed for uranium, plutonium, and thorium. The uranium content varied from 1.7 to 2.2 pc/g and thorium concentrations ranged from 0.016 to 0.021 pc/g. These values are similar to those obtained in other years. Plutonium concentrations were between 0.03 and 0.05 pc/g, and are attributed to fallout.

The total activities in samples collected from the reference sites are given in Table XXXVII. The results were very similar to those obtained earlier. As was noted for the on-site samples, the total beta activities in the off-site samples collected after September 1961 did not show any significant increase from the atmospheric nuclear testing during this period.

TABLE XXXVI

NONVOLATILE RADIOACTIVITY IN SURFACE SOIL NEAR ANL, 1960-61

Date Collected	Distance from ANL (miles)	No. of Samples	Alpha Activity (pc/g)		Beta Activity (pc/g)	
			Max	Av	Max	Av
April 28, 1960	10	3	19	17	54	46
	25	2	22	19	49	45
May 26, 1960	25	2	21	18	65	52
July 28, 1960	10	3	27	23	51	44
	25	2	31	27	62	55
August 17, 1960	10	2	28	23	82	72
August 31, 1960	10	3	27	18	59	43
	25	2	19	16	63	56
November 16, 1960	10	7	32	22	85	60
	25	6	26	20	59	48
1960	10	18	32	21	85	54
Summary	25	14	31	20	65	50
	10 and 25	32	32	21	85	52
March 30, 1961	10	2	21	18	49	46
April 27,28, 1961	10	4	27	24	61	54
	25	2	33	27	60	50
May 24, 1961	25	1	17	-	42	-
August 29, 1961	10	3	22	18	55	42
	25	2	22	22	54	52
September 21,22, 1961	10	4	22	14	51	33
	25	2	20	17	53	50
1961	10	13	27	19	61	44
Summary	25	7	33	21	60	51
	10 and 25	21	33	20	61	46

TABLE XXXVII

NONVOLATILE RADIOACTIVITY IN SURFACE SOIL AT REFERENCE SITES, 1960-61

Location	May 25-26, 1960		November 16-18, 1960		May 24-26, 1961		October 24, 1961	
	pcα/g	pcβ/g	pcα/g	pcβ/g	pcα/g	pcβ/g	pcα/g	pcβ/g
Oak Point State Park, Wisconsin	16	50	18	43	18	44	15	41
Delavan, Wisconsin	16	37	20	46	16	38	17	33
Starved Rock State Park, Illinois	25	38	18	34	20	32	17	29
Monticello, Indiana	16	44	19	44	20	38	14	29
St. Joseph, Michigan	14	40	5	19	7.8	21	8.0	17
Magician Lake, Michigan	12	50	14	30	11	30	17	55
Kankakee River State Park, Indiana	16	35	-	-	8.3	16	-	-
Average	16	42	16	36	14	31	15	34

F. Plants

Plant sampling was limited to grass because it was available at all locations, and intercomparison of results is more reliable if the same type of plants are collected at all locations. Total activities were

determined by counting ashed plant samples. The results are given in Tables XXXVIII, XXXIX, and XL in terms of the oven-dried sample.

TABLE XXXVIII

NONVOLATILE RADIOACTIVITY IN GRASS COLLECTED ON ANL SITE, 1960-61

Date Collected	No. of Samples	Alpha Activity (pc/g)		Beta Activity (pc/g)	
		Max	Av	Max	Av
April 29, 1960	3	0.99	0.50	52	41
July 26, 1960	1[a]	1.5	-	36	-
August 16, 1960	8[b]	0.77	0.39	54	28
August 19, 1960	1	0.33	-	35	-
November 18, 1960	6	0.81	0.54	45	27
1960 Summary	18	0.99	0.46	54	30
March 29, 1961	2	2.2	2.0	38	31
March 30, 1961	6	1.3	1.1	27	22
May 12, 1961	3	1.9	1.0	57	42
June 5, 1961	3	0.70	0.53	21	19
June 23, 1961	6[a]	21.5	4.6	90	53
June 28, 1961	1	0.52	-	37	-
August 29, 1961	1	1.0	-	27	-
September 19, 1961	6	1.1	0.6	64	39
1961 Summary	28	21.5	1.7	90	36

[a]Collected near uranium-storage shed.

[b]Collected near EBWR and CP-5 Reactor Buildings.

TABLE XXXIX

NONVOLATILE RADIOACTIVITY IN GRASS NEAR ANL, 1960-61

Date Collected	Distance from ANL (miles)	No. of Samples	Alpha Activity (pc/g)		Beta Activity (pc/g)	
			Max	Av	Max	Av
April 28,29, 1960	10	4	0.91	0.37	52	44
	25	2	0.42	0.40	46	43
May 26, 1960	25	2	0.24	0.17	29	28
July 17, 1960	10	2	0.21	0.17	60	50
September 29, 1960	10	3	1.1	0.68	35	28
	25	2	0.65	0.45	21	21
November 16,18, 1960	10	4	0.86	0.65	39	21
	25	4	1.4	1.3	34	26
1960	10	13	1.1	0.40	60	34
Summary	25	10	1.4	0.72	46	29
	10 and 25	23	1.4	0.54	60	32
April 3, 1961	10	2	1.1	0.9	30	22
	25	2	1.2	0.9	34	31
April 27, 1961	10	1	0.62	-	61	-
	25	1	1.7	-	17	-
May 24, 1961	25	1	0.72	-	39	-
August 29, 1961	10	3	0.82	0.49	52	33
	25	2	1.2	0.90	26	25
September 21, 1961	10	2	0.39	0.38	37	28
	25	3	0.45	0.42	56	37
1961	10	8	1.1	0.6	61	33
Summary	25	9	1.7	0.8	56	33
	10 and 25	17	1.7	0.7	61	33

TABLE XL

NONVOLATILE RADIOACTIVITY IN GRASS FROM REFERENCE SITES, 1960-61

Location	May 25-26, 1960		November 16-17, 1960		May 24-25, 1961		October 24-26, 1961	
	pcα/g	pcβ/g	pcα/g	pcβ/g	pcα/g	pcβ/g	pcα/g	pcβ/g
Delavan, Wisconsin	0.15	31	0.76	22	0.21	44	0.40	163
Oak Point State Park, Wisconsin	-	-	0.86	27	-	-	-	-
St. Joseph, Michigan	0.36	43	0.55	12	-	-	1.2	91
Magician Lake, Michigan	0.25	37	-	-	0.24	23	0.42	93
Starved Rock State Park, Illinois	0.29	37	1.1	20	0.63	46	0.89	193
Monticello, Indiana	0.08	23	1.1	27	-	-	0.66	90
Kankakee River State Park, Indiana	0.23	37	-	-	0.92	91	-	-
Average	0.23	35	0.87	22	0.50	51	0.71	126

The only abnormally high on-site total activity concentrations determined in this way were found in some of the samples collected near the uranium storage shed. The highest of these samples was collected on June 23, 1961. The bulk of the alpha activity in this sample was due to uranium. However, this grass also contained 0.64 pc of thorium and 0.14 pc of plutonium per gram, respectively, about ten and three times greater than the concentrations normally found in grass. The thorium activity was due to thorium-232 and thorium-228. The plutonium and thorium content of this area will be investigated further, but until additional information is available, it must be assumed that the plutonium and thorium in this sample, as well as the uranium, originated at Argonne.

At other locations, both on and off the site, except at the reference sites in October 1961, the total activities were normal and in the range found earlier. In October 1961, the reference-site samples contained about five times their normal beta activity. These samples contained short-lived fission products, and the increased beta activity was due to fallout from the nuclear testing conducted in the fall of 1961. Soil collected at the same time contained much smaller concentrations of fallout, indicating that grass is a much more sensitive fallout detector than soil.

Additional analyses on some of the samples gave the following results. Uranium concentrations varied between 0.015 and 0.15 pc/g, thorium concentrations between 0.004 and 0.08 pc/g, and plutonium concentrations between 0.006 and 0.05 pc/g. These values are in the range found during 1959. Cesium-137 concentrations were 0.1 to 0.2 pc/g, 10 to 50 times less than in 1959.

The concentrations of iodine-131 in soil and grass following the release beginning in February 1961 (see Section III-A) are given in Table XLI. The bulk of the iodine was released from a point in the southeast corner of square 12G (Figure 13). The concentrations in grass and soil decreased rapidly with distance from the point of release. Radioiodine was not detected in soil more than 150 yards from the point of release, while detectable amounts were present in grass 2400 yards from the release. The

concentrations in grass were approximately one hundred times greater than in soil from the same location, so that grass is a more sensitive indicator of surface iodine contamination than soil. Similar results were noted in the case of surface deposition of fallout from nuclear detonations. Small amounts of iodine-131 were detected in grass collected at the southern and western boundaries of the site, but none was detected near the northern and eastern periphery. This is consistent with the concentrations found in the off-site air samples, since in these samples iodine-131 was found only southeast and southwest of the site. These results indicate that the concept of prevailing westerly winds in this area cannot be relied upon to indicate the direction of travel of airborne contaminants.

TABLE XLI

IODINE-131 IN SURFACE SOIL AND GRASS, MARCH-JUNE, 1961

Location	Date	No. of Samples	Picocuries per gram at Collection Time			
			Surface Soil		Grass	
			Max	Av	Max	Av
ANL Site 12G	March 29	4	104	27	14.8×10^3	5.7×10^3
	May 11	4	-	-	11.4	7.3
	June 5	4	-	-	7.8	2.7
	June 28	1	-	-	<1	-
	July 26	2	-	-	<1	-
ANL Site 12H	March 29	4	28	10	1.8×10^3	0.86×10^3
	July 26	1	-	-	<1	-
ANL Site 11G	March 29	1	6.5	-	0.62×10^3	-
	July 26	1	-	-	<1	-
ANL Site 10H	March 29	1	<0.3	-	0.38×10^3	-
	June 5	4	-	-	7.8	2.7
	June 28	4	-	-	4.0	1.0
	July 26	3	-	-	<1	-
ANL Site F8	March 30	1	<0.3	-	-	-
ANL Site 11B	March 30	1	<0.3	-	5.0	-
ANL Site 15F	March 30	1	<0.3	-	<1	-
ANL Site 15L	March 30	1	<0.3	-	<1	-
ANL Site 12-O	March 30	1	<0.3	-	<1	-
ANL Site 6K	March 30	1	<0.3	-	4.9	-
ANL Site 12E	March 30	1	<0.3	-	23	-
Lemont	April 2	1	-	-	<1	-
Joliet	April 2	1	-	-	<1	-
Downers Grove	April 2	1	-	-	<1	-
Lombard	April 2	1	-	-	<1	-
Tinley Park	April 2	1	-	-	<1	-

Because of the relative amounts of iodine-131 found in soil and grass on the site, off-site sampling for surface iodine was limited to grass. As indicated in Table XLI, iodine-131 was not detected in any of the off-site grass samples. Although iodine-131 was found in some air samples off the site, the concentrations apparently were too low to result in the deposition of detectable amounts on the grass. In this respect, the absence of iodine-131 in grass from Lemont, only 2.5 miles southwest of the point of release, should be noted. The rate of disappearance of iodine-131 from grass was appreciably greater than the 8-day half-life, as would be expected for a variety of chemical and physical reasons. Iodine-131 from this release was not detected in grass after June 1961. These results, together with the low concentrations found in off-site air samples, indicate that the iodine released by Argonne did not constitute a health hazard.

Since air, soil, and grass contained iodine-131, it was of interest to determine if iodine-131 could also be detected in animal life in the area. A fox and skunk collected in March 1961 near the point of major iodine-131 release contained detectable amounts (25-50 pc/g) of iodine-131 in their thyroids. Radioiodine could not be detected in fish obtained from several lagoons on the site during this period.

G. Site A Samples

A former portion of Argonne National Laboratory, known as Site A, was located about 4 miles east of the present site. An area of about 16,000 ft^2 at Site A was used for the burial of contaminated waste material. In 1954 as much as possible of this waste was removed and the area covered with a concrete cap, one foot thick and eight feet deep, along the sides. The area is drained by an intermittent stream, and samples of water and bottom silt were collected from this stream in 1954 to determine if activity had been leached from the burial area. Soil borings were also taken on the downstream side of the burial area for the same purpose. No contamination from the burial area was found in any of the samples. The results are given in ANL-5446.

The same locations were resampled in August of 1960 and 1961. Water was not present in the stream at the times of sampling. The soil borings in 1960 and 1961 contained from 22 to 35 pcα/g and from 52 to 68 pcβ/g, compared to activities of 19 to 33 pcα/g and 50 to 86 pcβ/g in 1954. Thus, no increase in activity was detected adjacent to the burial site. Additional sampling of the area will be done periodically.

Figure 1

AVERAGE RADIOACTIVITY IN SURFACE WATER, 1960

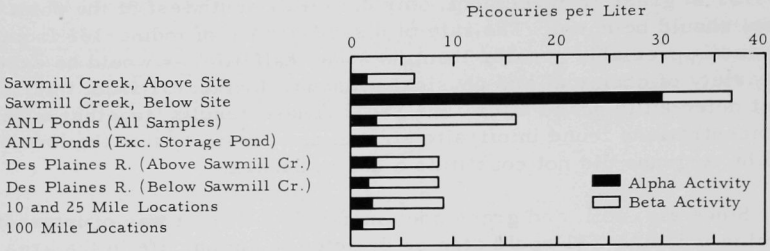


Figure 2

AVERAGE RADIOACTIVITY IN BOTTOM SILT, 1960

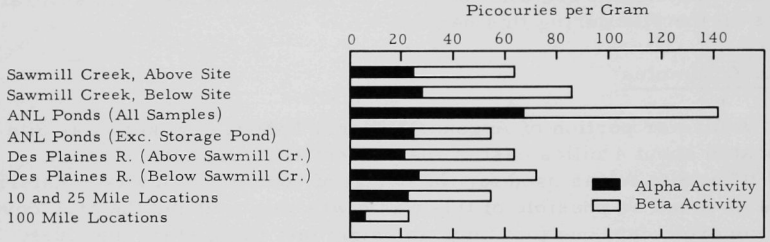


Figure 3

AVERAGE RADIOACTIVITY IN SOIL AND PLANTS, 1960

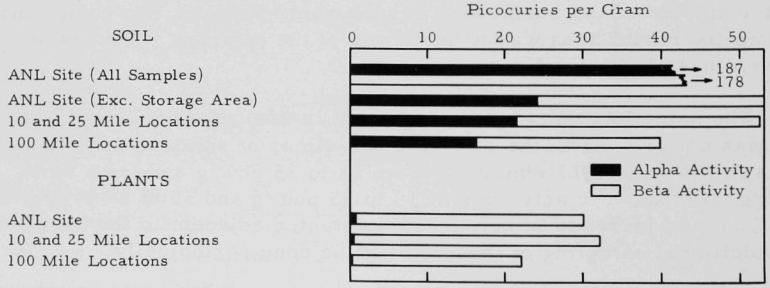


Figure 4

AVERAGE RADIOACTIVITY IN SURFACE WATER, 1961

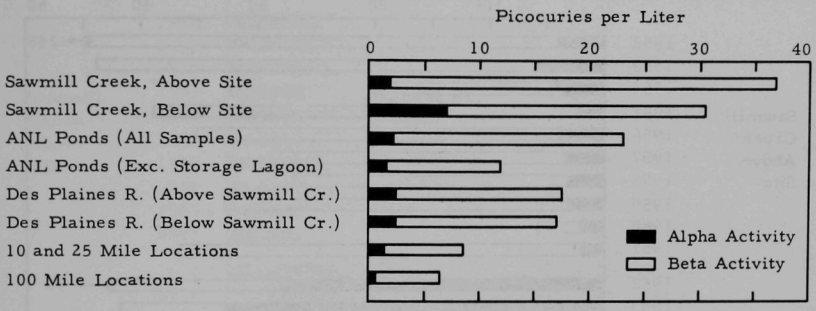


Figure 5

AVERAGE RADIOACTIVITY IN BOTTOM SILT, 1961

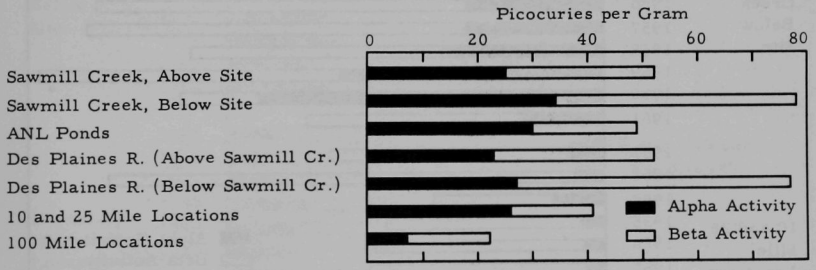


Figure 6

AVERAGE RADIOACTIVITY IN SOIL AND PLANTS, 1961

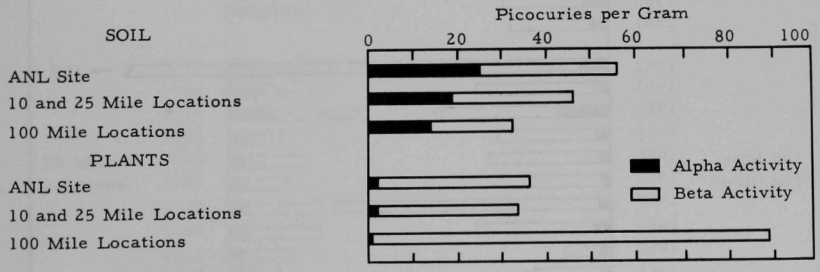


Figure 7

AVERAGE RADIOACTIVITY IN SURFACE WATER, 1952-61

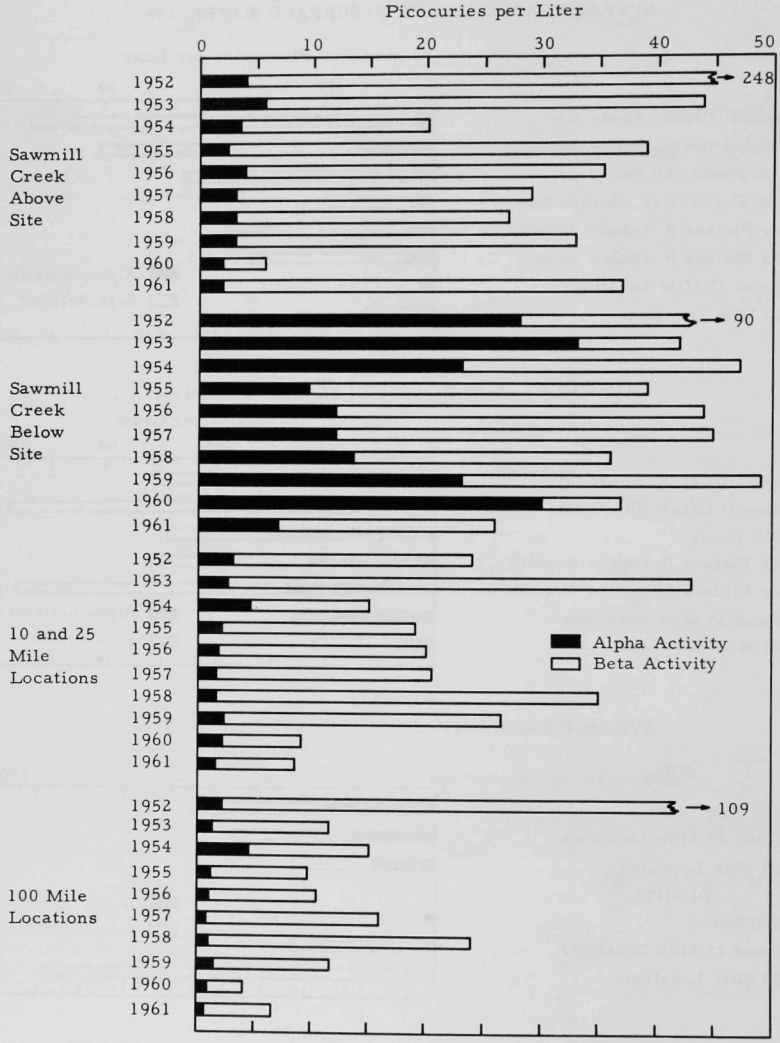


Figure 8

AVERAGE RADIOACTIVITY IN BOTTOM SILT, 1952-61

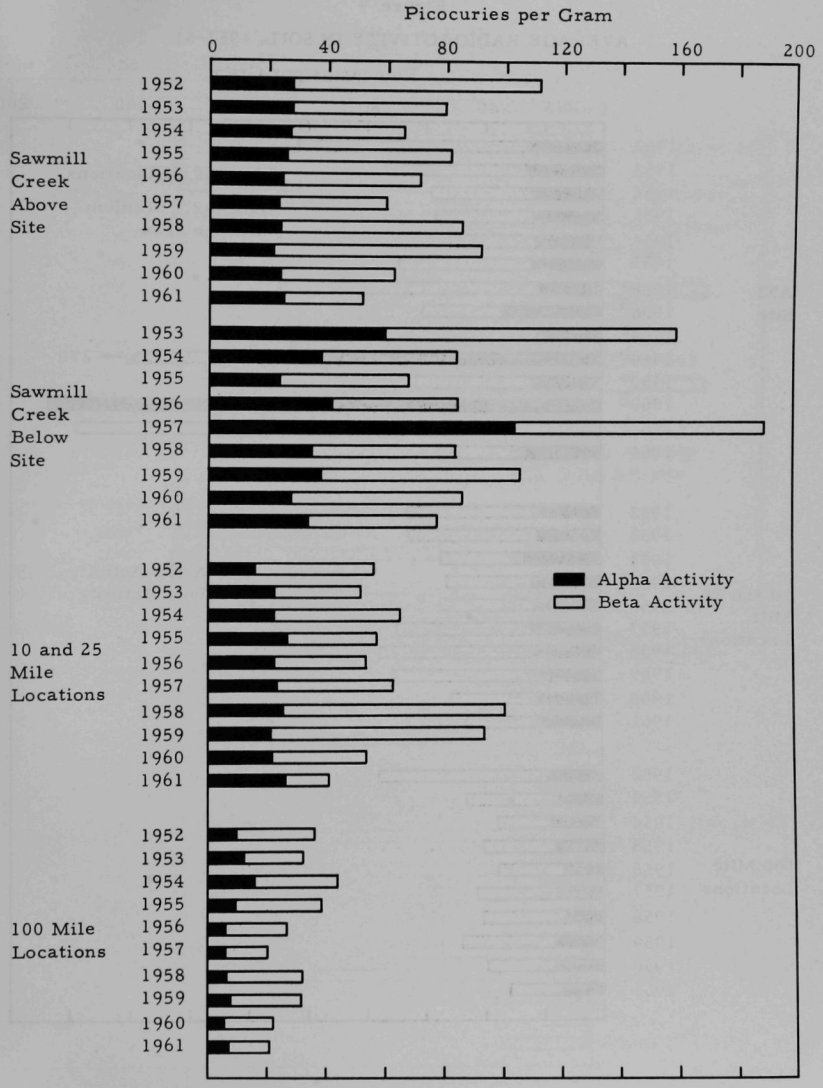


Figure 9

AVERAGE RADIOACTIVITY IN SOIL, 1952-61

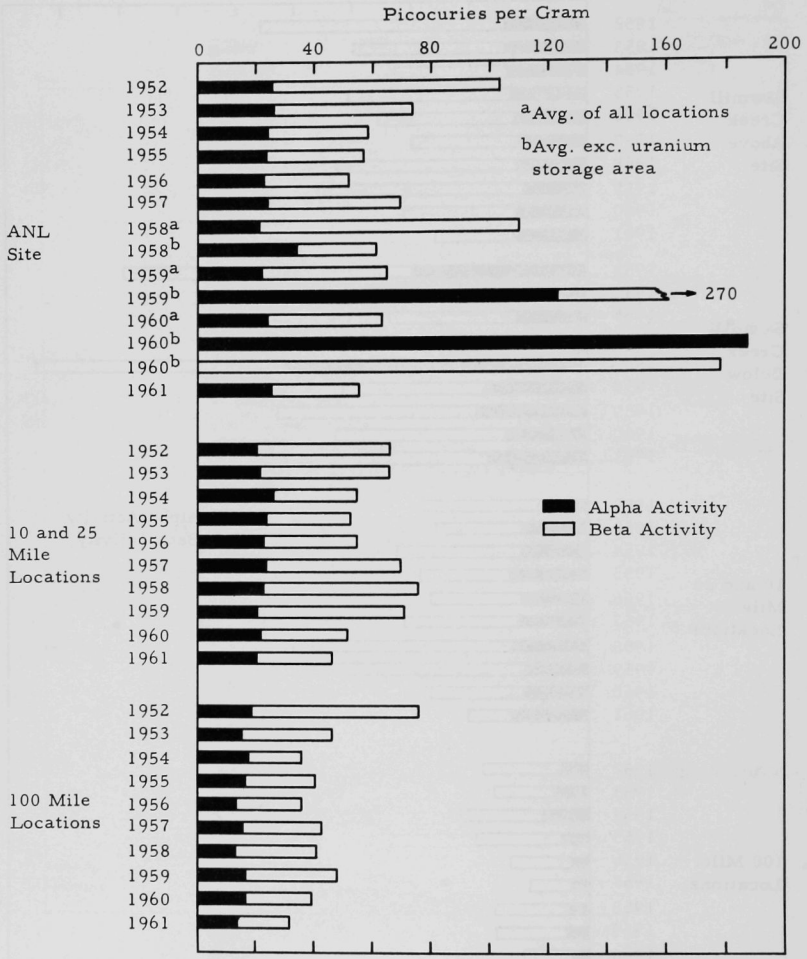


Figure 10

AVERAGE RADIOACTIVITY IN PLANTS, 1952-61

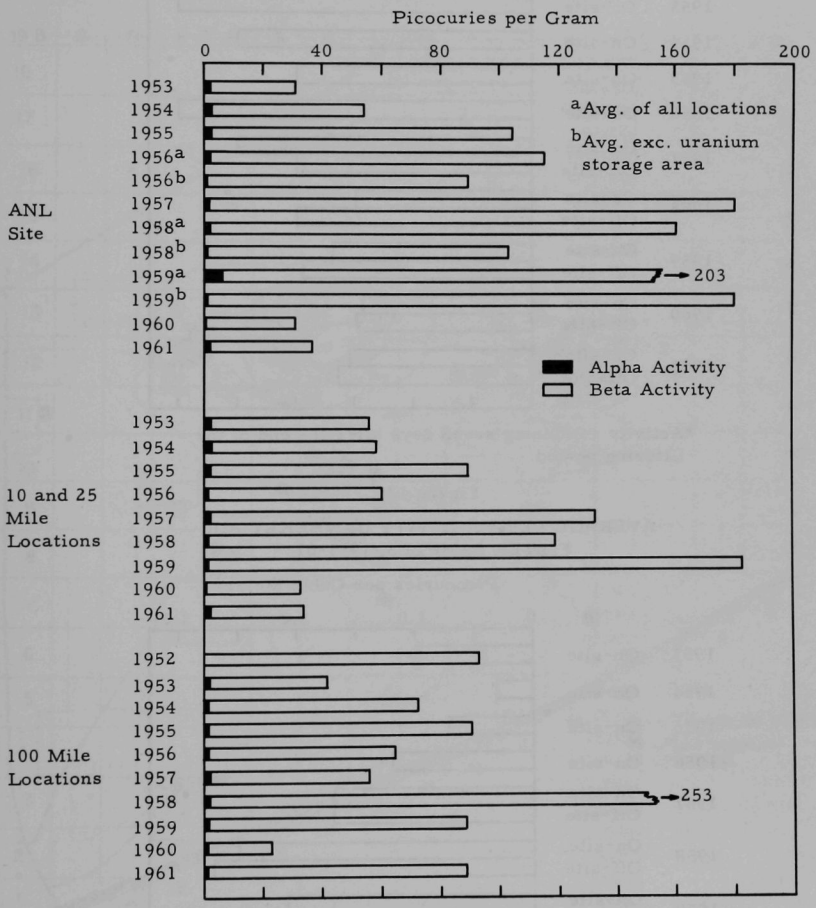
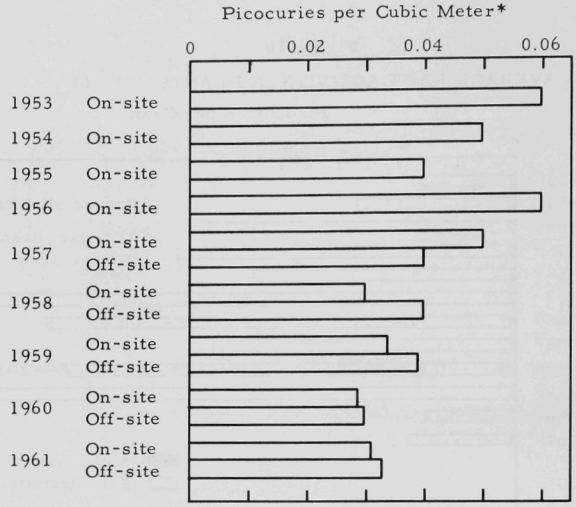


Figure 11

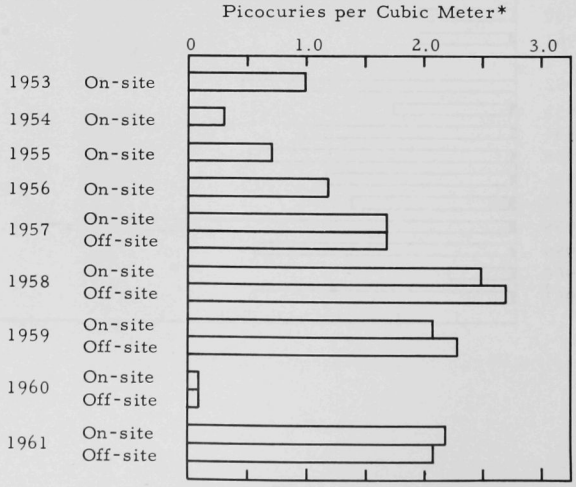
AVERAGE ALPHA ACTIVITY IN WEEKLY AIR FILTER SAMPLES, 1953-61



*Activity remaining seven days after the end of the filtering period.

Figure 12

AVERAGE BETA ACTIVITY IN WEEKLY AIR FILTER SAMPLES, 1953-61



*Activity remaining seven days after the end of the filtering period.

Figure 13

SAMPLING LOCATIONS ON THE SITE OF
ARGONNE NATIONAL LABORATORY

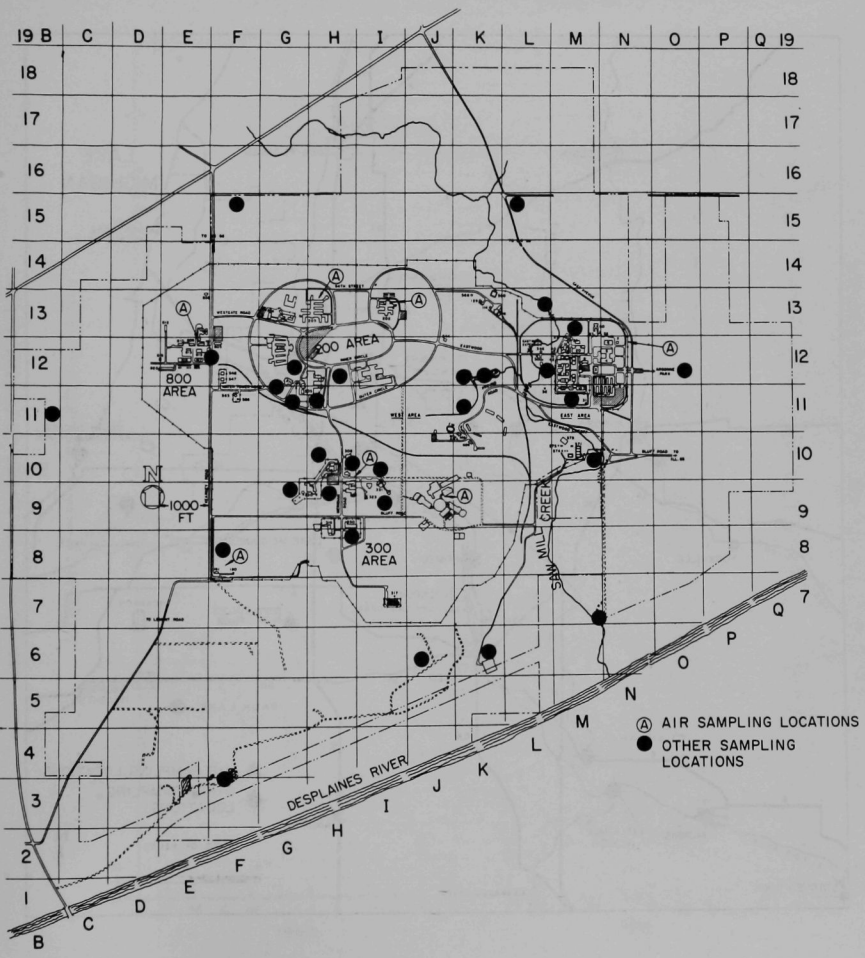


Figure 14

SAMPLING LOCATIONS NEAR ARGONNE NATIONAL LABORATORY

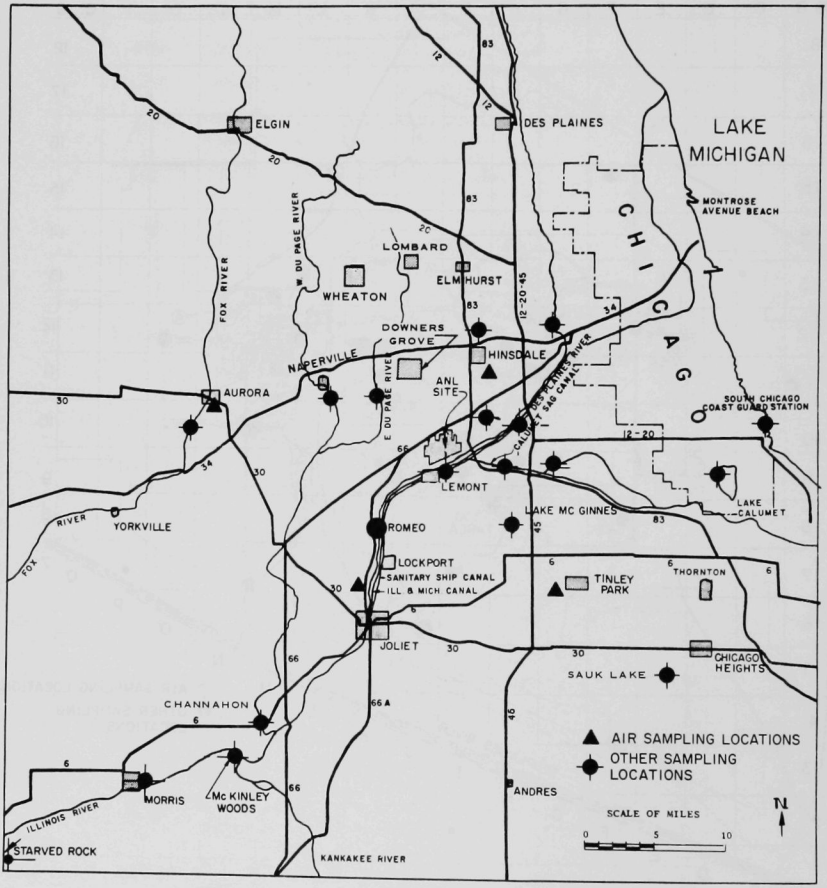


Figure 15

TOTAL BETA ACTIVITY IN PRECIPITATION AND AIR FILTER SAMPLES, 1960-61